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VISIBLE AND INFRARED LASER-INDUCED DAMAGE TO TRANSPARENT MATERIALS

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13. ABSTRACT

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Damage from inclusions was observed in various dielectric coatings. It was found that inclusions having diameters ~3.5 μm were responsible for damage from Q-switched pulses and that inclusions with diameters less than ~ 0.4 μm were responsible for threshold damage from 20 ps mode-locked pulses.

An operating CO_2 TEA laser has been assembled for use as a source for damage measurements. Future cavity refinements are needed to restrict lasing to a TEM_{00} mode with peak powers in excess of about 150 KW.

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TECHNICAL REPORT SUMMARY

Laser-induced damage to initially transparent materials represents a serious limitation to the design and operation of high power lasers. In order to assess these limitations and to provide standards for material performance, it is necessary to obtain reliable measurements of damage intensities. The primary goal of the present program is to measure damage intensities in various transparent solids and to interpret these results in terms of possible damage mechanisms. Using techniques which we developed in earlier work, we have been able to isolate and to study the intrinsic damage process of electron avalanche breakdown. Such work establishes measured upper limits for the propagation of high intensity light in solids and indicates how these intrinsic limits change with laser pulse and material characteristics. We have, in addition, observed damage from small absorbing inclusions and have begun to develop techniques which can be applied to optical materials evaluation. As part of the present effort, a CO_2 TEA laser is being designed and constructed as a TEM_{00} mode, 10.6 μm source for optical damage.

A study of intrinsic damage in polycrystalline and disordered materials has been completed and interpreted in terms of electron avalanche breakdown. By comparing the optical bulk damage fields for a polycrystal, various single-crystal alloys, and two glassy solids to the damage fields for the respective pure single crystals, it was found that only severe lattice disorder such as present in completely amorphous fused quartz causes the damage fields to increase. For the polycrystal and the less disordered systems, the damage fields are the same as those of the pure single crystal.

Measurements have been made of the pulse width independence of intrinsic damage in NaCl at 1.06 μm. It was found that the intrinsic damage field increased by nearly an order of magnitude to over 10^7 volts/cm as the laser pulse duration was decreased from 10 ns to 15 ps. These results, interpreted in terms of electron avalanche breakdown, are the first measurements of intrinsic damage induced by mode-locked laser pulses.

Finally, optical damage to dielectric coatings was studied with weakly focused ruby laser pulses having durations between 20 ns and 20 ps. Under these conditions of measurement, damage was found to result from highly absorbing inclusions. Inclusions with diameters $\gtrsim$ 3.5 μm were most easily damaged with 20 ns pulses while inclusions with diameters $\lesssim$ 0.4 μm determined damage resistance to 20 ps pulses.

The present work on intrinsic damage has extended over knowledge of electron avalanche breakdown. It may now be possible to develop realistic models of this damage process that will enable us to accurately predict the effects of material parameters on intrinsic damage fields at least at red and near infrared frequencies. Damage measurements should be extended to higher optical frequencies in order to determine if damage from electron avalanche breakdown still dominates as the material bandgap is approached.

Our studies of inclusion and self-focusing damage can serve as a model for future materials evaluation. Since inclusions and self-focusing normally limit the intensity of light which materials can withstand, such evaluation is essential for the development of more damage-resistant optics and for the design of damage resistent systems. In future work we will consider standardized techniques for evaluating inclusion content in laser materials and for measuring self-focusing parameters.

ABSTRACT

By using Q-switched and mode-locked Nd:YAG lasers, we have measured the dependence of intrinsic damage fields on both lattice disorder and laser pulse duration and have interpreted the results in terms of avalanche breakdown. It was found that severe lattice disorder such as present in fused silica measurably increases the intrinsic damage intensity. The breakdown field was also found to increase when the pulse duration was reduced below about 1 ns.

Damage from inclusions was observed in various dielectric coatings. It was found that inclusions having diameters $\sim 3.5\ \mu m$ were responsible for damage from Q-switched pulses and that inclusions with diameters less than $\sim 0.4\ \mu m$ were responsible for threshold damage from 20 ps mode-locked pulses.

An operating CO_2 TEA laser has been assembled for use as a source for damage measurements. Future cavity refinements are needed to restrict lasing to a TEM_{00} mode with peak powers in excess of about 150 KW.

FOREWORD

This scientific report describes work performed under Contract No. F19628-73-C-0127 between 1 January 1973 and 30 June 1973. The report was assigned a Raytheon internal number S-1593.

Work was carried out at the Raytheon Research Division in Waltham, Massachusetts, the Naval Research Laboratories in Washington, D.C., and the Air Force Cambridge Research Laboratories in Bedford, Mass. The Principal Investigator until June 30, 1973 was Dr. Michael Bass. After that date Dr. David Fradin became Principal Investigator. This technical report was prepared under the direction of Dr. Fradin in collaboration with Dr. Bass and D.P. Bua. The authors have benefitted from the many discussions with Keimpe Andringa, Dr. Frank A. Horrigan, Dr. Thomas Deutsch and Dr. Robert Rudko. Miss Carol Christian assisted in the ZnSe study. Dr. A. Linz generously provided several of the samples used in the damage study of disorder materials and J. VanderSande conducted electron reflection diffraction measurements for that study.

This report was submitted by the authors on 31 July 1973.

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I. GENERAL INTRODUCTION

The objective of this program is to analyze and study data obtained from carefully controlled damage measurements at wavelengths between 0.69 μm and 10.6 μm and at various laser pulse durations. Laser damage thresholds of various materials of interest will be determined using techniques developed during the past year, and the results will be analyzed to ascertain the role of electron avalanche breakdown as the intrinsic damage process. Theoretical evaluations of electron avalanche breakdown will be conducted. Techniques for the measurement of self-focusing parameters using optical damage will be developed and applied to various materials.

In the first half of the program our efforts were split into four major areas. They were: (1) design and assembly of a pulsed CO_2 TEA laser for use in the 10.6 μm damage measurements, (2) a study of laser damage in crystalline and amorphous forms of the same material to determine the effects of local disorder on intrinsic damage processes, and (3) measurements of the dependence of breakdown fields on laser pulse duration.

The CO_2 laser has been designed and constructed. By using Rogowski shaped electrodes, it has been possible to obtain a uniform glow discharge inside the resonator and to produce approximately 0.7 joules of multimode laser output. Refinements in the design of the cavity optics to produce single mode output are in progress.

We have demonstrated that severe disorder such as present in a highly amorphous material can increase the intrinsic damage field in a manner consistent with an electron avalanche breakdown damage mechanism. The intrinsic breakdown strengths of polycrystals and alloys, on the other hand, are not measurably affected by lattice disorder.

In addition to the above work which was carried out at the Raytheon Research Division, a joint effort between Raytheon, Harvard University

and Naval Research Laboratories personnel measured the pulse duration dependence to intrinsic laser damage in NaCl. Facilities of the Naval Research Laboratories were used for this work. The results of this study were interpreted in terms of electron avalanche breakdown and compared to previously published measurements of dc dielectric breakdown in thin samples of NaCl.

A second joint effort, conducted with Air Force Cambridge Reseach Laboratories personnel at the AFCRL's Optical Physics Laboratory, studied optical damage to dielectric coatings by the use of weakly focused ruby laser pulses of varying duration. Threshold damage was found to result from inclusions whose diameter varied in a predictable manner with pulse width.

Appendices A and B contain the abstracts of papers given this year at the American Physical Society Winter Meeting in New York City (January) and at the Twelfth Symposium on Electron, Ion and Laser Beam Technology at the Massachusetts Institute of Technology (May). A paper presented at the Fifth Laser Damage Symposium in Boulder, Colorado, in May, 1973, is reproduced in Appendix C.

II. DESIGN AND CONSTRUCTION OF A CO_2 TEA LASER

A. Introduction

A major objective of the present program has been the design and construction of a CO_2 transverse-excited-atmospheric (TEA) laser. This work has been divided into three phases:

1. selection of a gas discharge design
2. construction of an operating laser resonator
3. application of design refinements of the resonator optics to produce TEM_{00} output with sufficient power and spectral purity to perform optical damage measurements.

The first two phases were completed during the first semi-annual period. A double Rogowski electrode configuration[1] was selected for our use. Using a published electrical design[2] with minor modifications, we were able to achieve a stable, uniform discharge, and, in initial measurements, to produce approximately 0.7 joules of multimode output at 10.6 μm.

B. Resonator Design

High peak powers can be obtained with CO_2 lasers by use of atmospheric pressure and fast electric discharges.[3] There are two reasons for using transverse excitation at high pressures instead of a longitudinal discharge. First, lower voltages are required to produce gas breakdown since the discharge path is reduced. In addition, the discharge impedance is lower for transverse excitation so that energy can be rapidly injected into the discharge volume. This second advantage is important because it allows the laser to be excited in times short compared to the excited state lifetime of the CO_2 molecule, a lifetime equal to about 10 μs at atmospheric pressure.[4] As a result of this rapid excitation, the laser is Q-switched without using such intracavity devices as electro-optic shutters or rotating mirrors.

A number of schemes for transverse excitation have been discussed in the literature.[1-3,5,6] Two of them, pin electrodes and Rogowski electrodes with trigger wires, were compared in our laboratory. The latter design was chosen because it offered the advantages of a large discharge volume, comparative simplicity, and a relatively low level of radiated rf noise.

The laser was constructed with two parallel graphite electrodes shaped to approximate a Rogowski profile.[7] Two thin (9-mil) tungsten trigger wires running parallel to the electrodes and placed just outside the lasing region (see Fig. 1) provide preionization of the discharge volume[1] with the main discharge occurring across the graphite electrodes.

Chang and Wood[2] have published a simple electrical arrangement for this type of laser, and we have adopted their design with minor modifications. Figure 2 shows a schematic of this design with the circuit element values that we have used in our system. To produce the discharge, capacitors C_1 and C_2 are initially charged to about 30 kV. A short voltage spike from a trigger transformer fires spark gap SG and current i_1 begins to flow. The spark gap is a commercial device (Tachisto) especially modified for our use.[8] Because of the spark gap inductance, i_1 starts to oscillate and to drive the trigger wires T and cathode K to negative voltages. When the voltage at T begins to drop below ground, an arc discharge is produced between T and the resonator anode A. UV light from this arc causes preionization in the gas volume between the laser electrodes K and A and helps to initiate a large-volume uniform main discharge. This discharge rapidly damps out the ringing current and absorbs most of the energy stored in the circuit.[2]

The discharge characteristics are influenced by the gases flowing in the resonator. We have used approximately 800 torr of N_2:CO_2:He mixed in the ratio 0.8:1:5.5. This mixture produces both a uniform glow discharge and optimized laser output. No vacuum pump is used in this system.[2] Instead, the gases are passed through an exhaust in the resonator box at a total rate of about 33 liters/minute.

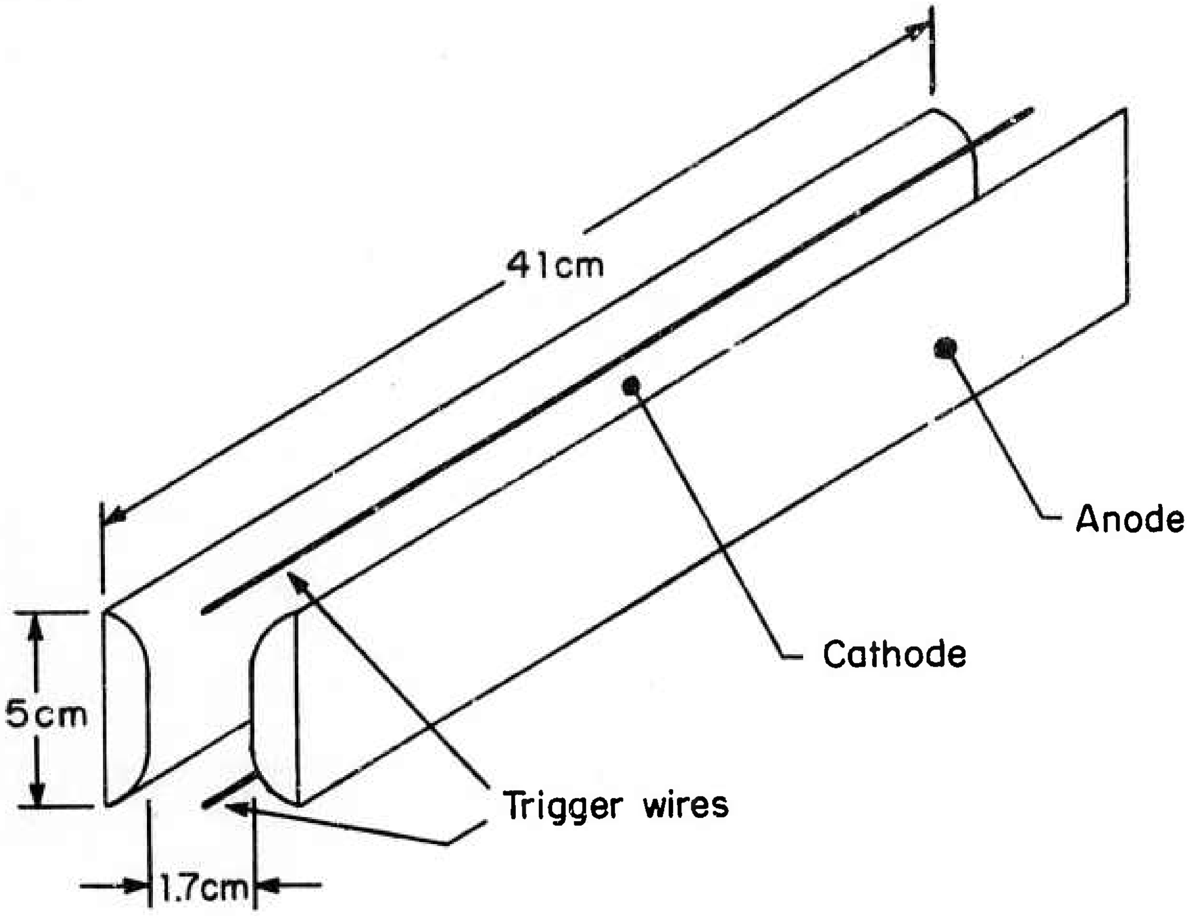
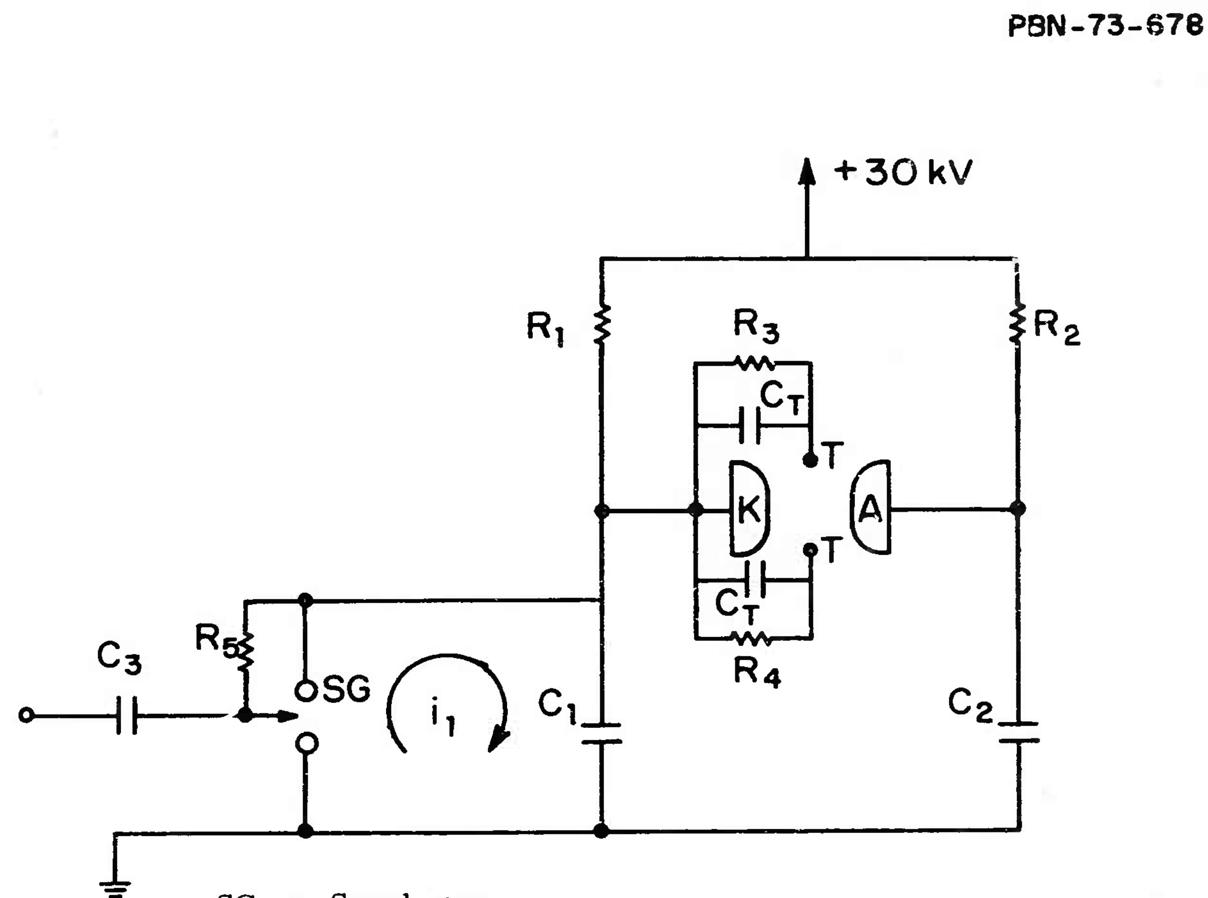


Fig. 1 Rogowski Shaped Graphite Electrodes. Each electrode polished to remove sharp edges. The trigger wires are 9 mil tungsten.



SG = Spark gap

R_1 = R_2 = 75kΩ, 100 W

R_3 = R_4 = 1kΩ, 2 W [(2) 500 Ω, 2 Watt resistors in series]

R_5 = 10 MΩ High voltage

C_1 = C_2 = 0.0432μf [(12) 3600 pf, 30 kV Sprague capacitor]

C_3 = 500 Pf

Fig. 2 Schematic of Electrical Design of the Laser Discharge (see Ref. 2).

The physical design of the resonator is also modeled after Ref. 2. Figure 3 is a sketch of the resonator showing the placement of the charging capacitors and spark gap and the design of the electrode mounts. A photograph of the actual laser is reproduced in Fig. 4. Because of the use of thick resonator walls, the present laser can be operated with other gases at well below atmospheric pressure.

Initial tests on this resonator produced about 0.7 joules of laser output at 10.6 μm with peak powers of about 0.6 Mw. Mirror reflectivity had not been optimized nor had the light beam been apertured for transverse mode selection. Firing at about 1 pps, the pulse energy stability was about 5 percent.

C. Future Optical Design of the Resonator

Performance requirements for the CO_2 laser are determined by its intended use as a light source for optical damage experiments. It is essential for our purposes that the laser operate in a TEM_{00} mode and that it achieves peak powers in excess of about 150 KW. It is highly desirable that the laser output contain a single longitudinal mode or that it be made completely time-resolved as in the work of Yablonovitch.[9] Spectral purity is not essential because the effects of high-frequency mode beating tend to average out when the mechanism is either electron avalanche breakdown[10] or inclusion absorption.[11]

A number of optical configurations can be used to produce TEM_{00} mode operation. The simplest is a resonator having mirrors with radii of curvature larger than the resonator length and having an intercavity aperture constraining the cavity Fresnel number. Because this configuation restricts the mode volume and hence the available energy output, it may be necessary to use lenses inside the cavity to increase the mode size[12] or to design an unstable resonator with annular output coupling.[13]

Longitudinal mode selection, if necessary, will be accomplished with an intracavity etalon aligned normal to the cavity axis.[14,15] This

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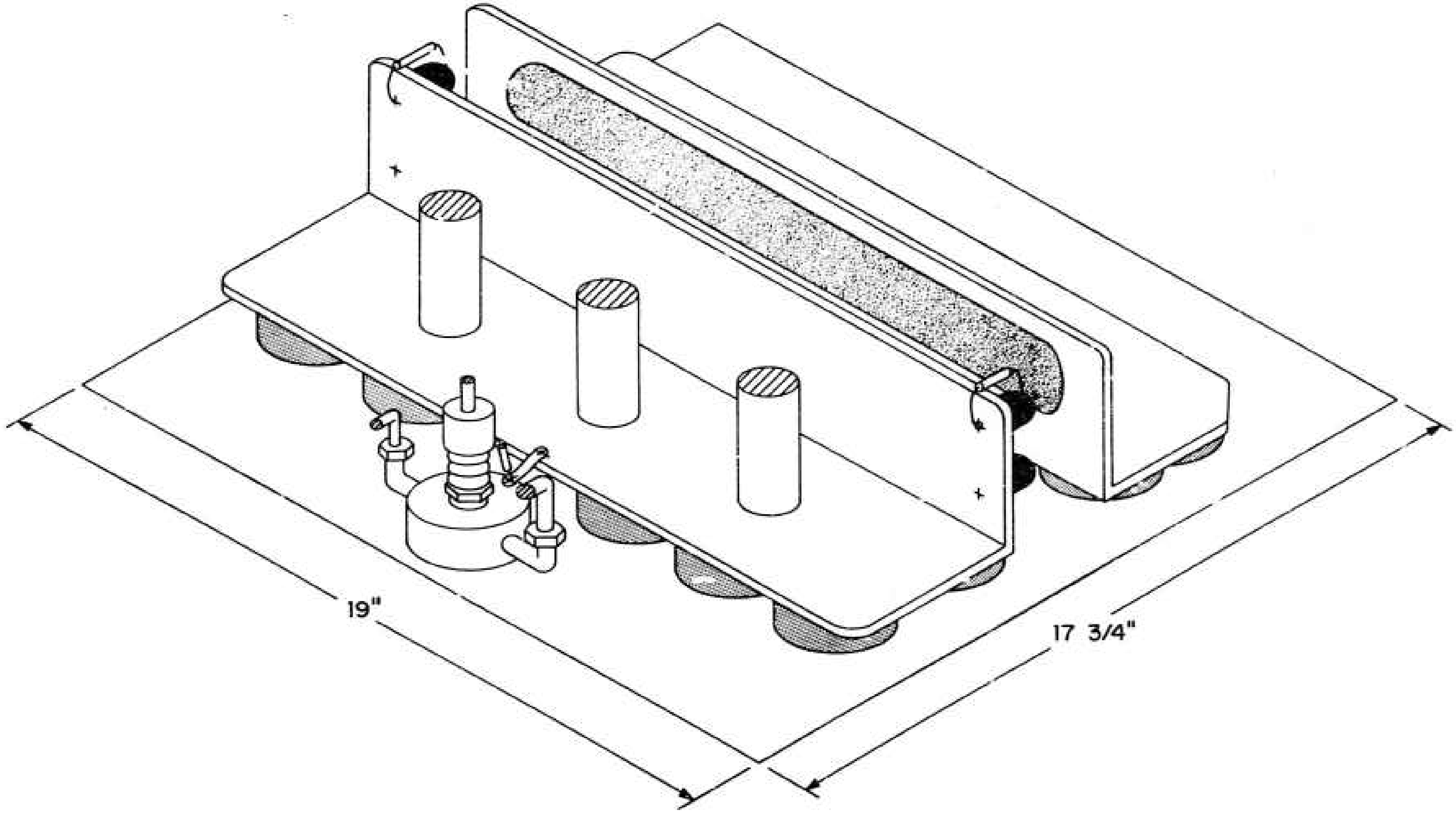
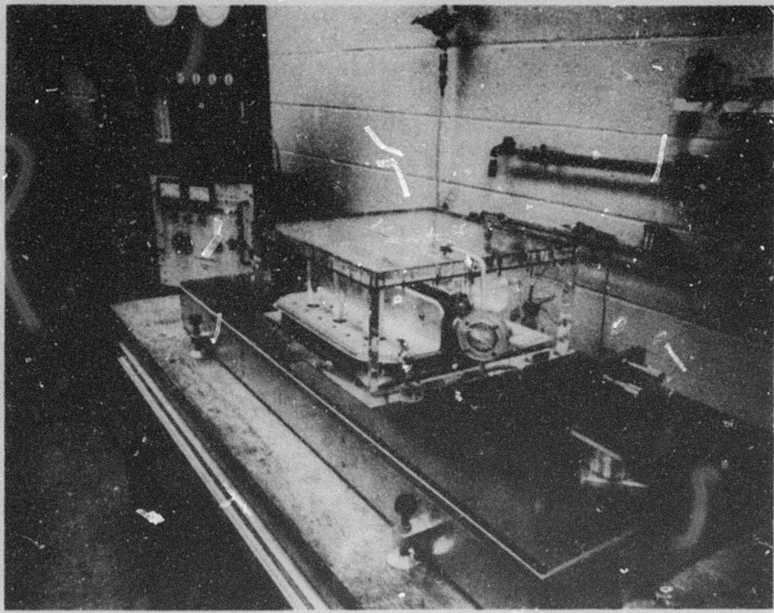


Fig. 3 Pictoral View of CO_2 Laser with Discharge Circuit.



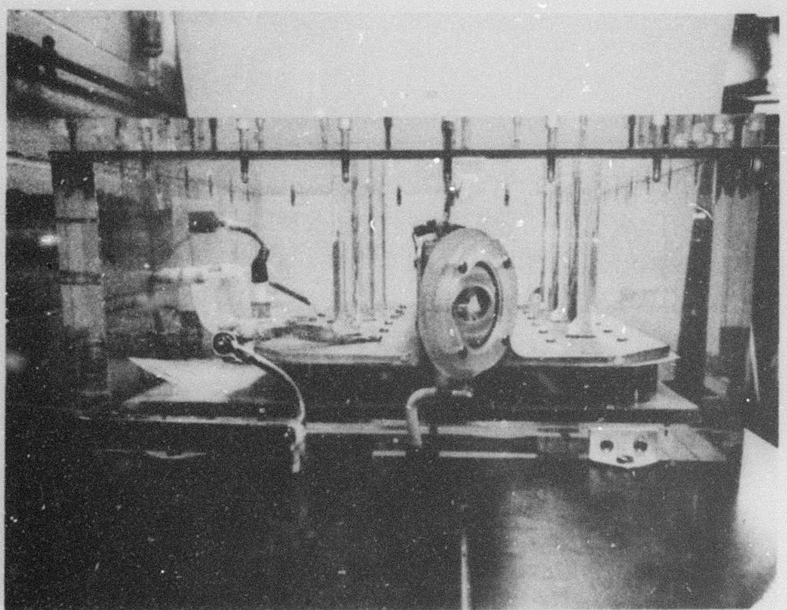


Fig. 4 Two Views of the Actual CO_2 Laser Built for this Program.

same technique has been used to restrict the number of lasing modes in ruby and Nd:YAG lasers where an etalon was formed by the highly reflecting end mirror and the plane, antireflection-coated front surface of the laser rod.[14] The high gain of the medium enhanced the effective reflectivity of the rod face sufficiently to produce an effective etalon with a finesse of about 6 and a free spectral range comparable to that of the total resonator. In this manner the number of longitudinal modes was restricted to four or less. In CO_2 TEA lasers, an uncoated germanium flat positioned between the discharge region and the output mirror produces similar spectral selection.[15]

Pulsed CO_2 lasers may operate on more than one rotational line. Since the lines of the P branches of either rotational band are separated by about 50 GHz, simultaneous lasing on more than one rotational line will produce ultrahigh frequency beating. As already noted, such fast time structure should not affect the results of optical damage measurements. For this reason no special attempt will be made to restrict lasing to a single rotational branch.

D. Conclusions

We have completed the design and construction of an operating CO_2 TEA laser. Before this system can be used as a light source for optical damage experiments, the cavity optics must be designed to produce TEM_{00} output with peak powers in excess of 150 KW. High spectral purity, though not essential, can be achieved through the use of an intracavity etalon.

III. EFFECTS OF LATTICE DISORDER ON THE INTRINSIC OPTICAL DAMAGE FIELDS OF SOLIDS

A. Introduction

Intrinsic damage fields have previously been determined only for pure single crystals. Recent interest has developed, however, in disordered solids as possible new materials for high power laser optics. Disordered solids, particularly polycrystals and alloys, are attractive because of their superior physical properties. In addition, certain materials such as ZnSe are more easily produced in polycrystalline form than as single crystals. There are, then, practical reasons for measuring the intrinsic breakdown fields of disordered solids.

A more fundamental reason for such measurements concern the nature of the intrinsic damage process. It has been demonstrated that electron avalanche breakdown is responsible for intrinsic damage of transparent solids. Since our previous work had established that effective electron collision times are on the order of 10^{-16} to 10^{-15} sec, it appears that the hot electrons involved in the avalanche are changing their moments rapidly by effectively colliding with every atom in their paths. This fact implies that only severe lattice disorder should affect the avalanche. It is argued in Sec. B, in fact, that the intrinsic damage field does not change unless disorder exists on the scale of 10 atomic units or less. If such severe disorder exists, it will be more difficult to heat the electron population and hence the damage field will rise. A measurement of the intrinsic damage in disordered materials, therefore, is a test of our physical conception of electron avalanche breakdown.

We have measured the intrinsic optical damage fields of three systems having different degrees of disorder - polycrystal KCl, single-crystal KCl-KBr alloys, and glass solids. It was found that extreme lattice disorder such as present in a highly amorphous material such as fused quartz causes the damage field to increase whereas less severe disorder does not measurably affect the breakdown strength. This result is consistent with our predictions based on avalanche breakdown as being the damage mechanism. It demonstrates, in addition, that the intrinsic limits for light propagation in single crystals and polycrystals of the same materials are identical.

B. Effects of Lattice Disorder on the Intrinsic Optical Damage Fields of Solids

Measurements of the effects of lattice disorder on the intrinsic laser-induced damage fields of transparent solids are reported. Extreme lattice disorder such as present in a highly amorphous material such as fused quartz causes the damage field to increase whereas less severe disorder does not measurably affect the breakdown strength. This is consistent with an electon avalanche intrinsic damage mechanism.

Studies of dc dielectric breakdown have shown that severe lattice disorder can increase the electric strength of solids.[16] Since it has recently been observed that intrinsic optical damage in transparent solids appears to develop from the same mechanism that is responsible for the dc dielectric breakdown,[9,10,17] severe lattice disorder should also increase the optical damage fields of solids.[9]

Measurements are reported in this paper of optical bulk damage in three systems having different degress of disorder -- polycrystal KCl, single-crystal KBr-KCl alloys, and glassy solids. In each case the damage field for the disordered system was compared to the optical strength of the corresponding crystal. It was found that severe lattice disorder such as present in the completely amorphous system (fused quartz) causes the intrinsic optical damage fields to increase. This observation is consistent with dc breakdown experiments[16] and with simple models of electron avalanche breakdown.[19]

The laser system and the experimental techniques used here are described in Ref. 10. A Q-switched, TEM_{00} Nd:YAG laser was used to induce damage inside the bulk of various samples. Self-focusing was avoided by restricting the laser input power,[9,10] and damage from inclusions was distinguished from intrinsic damage by examining both the morphology of the damage sites[9,10] and the temporal shape of light pulses transmitted through the sample.[18] Only data obtained from intrinsic damage events were considered in the present work. The damage field was defined as that value of on-axis, root-mean-square electric field necessary to induce damage on a single shot with a probability of 0.5.[10,20]

The damage field of the large-grain (20 μm) polycrystal was the same as that measured in the single crystal, and the damage fields for the alloys were intermediate between the damage fields of the constituents. In quartz, on the other hand, the disordered form was noticeably stronger than the crystal, the ratio of damage intensities being 5 ± 1. This ratio is identical to the corresponding ratio of surface damage fields measured in previous work.

Damage data were taken on a second amorphous-crystal pair, $Ba_2MgGe_2O_7$ (BMGO). Several measurements had indicated that the BMGO glass was distorted. An X-ray Laue pattern of the glass gave no evidence of crystallinity. Similarly, the directions of the stress-induced fractures that normally accompany optical damage[10] (Fig. 5) were random in the amorphous BMGO as expected for a glass, whereas fracture in the crystal occurred along well-defined crystal directions. Disorder on a local scale in BMGO glass was evident in the luminescence studies of Munasinghe and Linz.[21] When our sample was carefully studied with electron reflection diffraction,[22] however, it was found that regions ($\gtrsim$ 2000Å) of crystal ordering existed. Within the volume irradiated by the laser, therefore, the BMGO glass was not completely amorphous. It was actually a mixed crystal-amorphous phase. For the BMGO system, the damage fields of the glass and the crystal were the same within experimental error.

Table I and Fig. 6 summarize the measured optical damage fields.

It is to be expected that the large-grain polycrystal should have the same damage field as the single crystal. The average grain (20 μm) and the laser focal volume were comparable so that in the high intensity region near the beam axis where breakdown is observed to initiate, the sample looks like a single crystal.

By a simplified argument we can estimate the degree of disorder necessary to affect the breakdown strength. Classical theories of avalanche breakdown[19] predict that the dynamics of electrons with energies

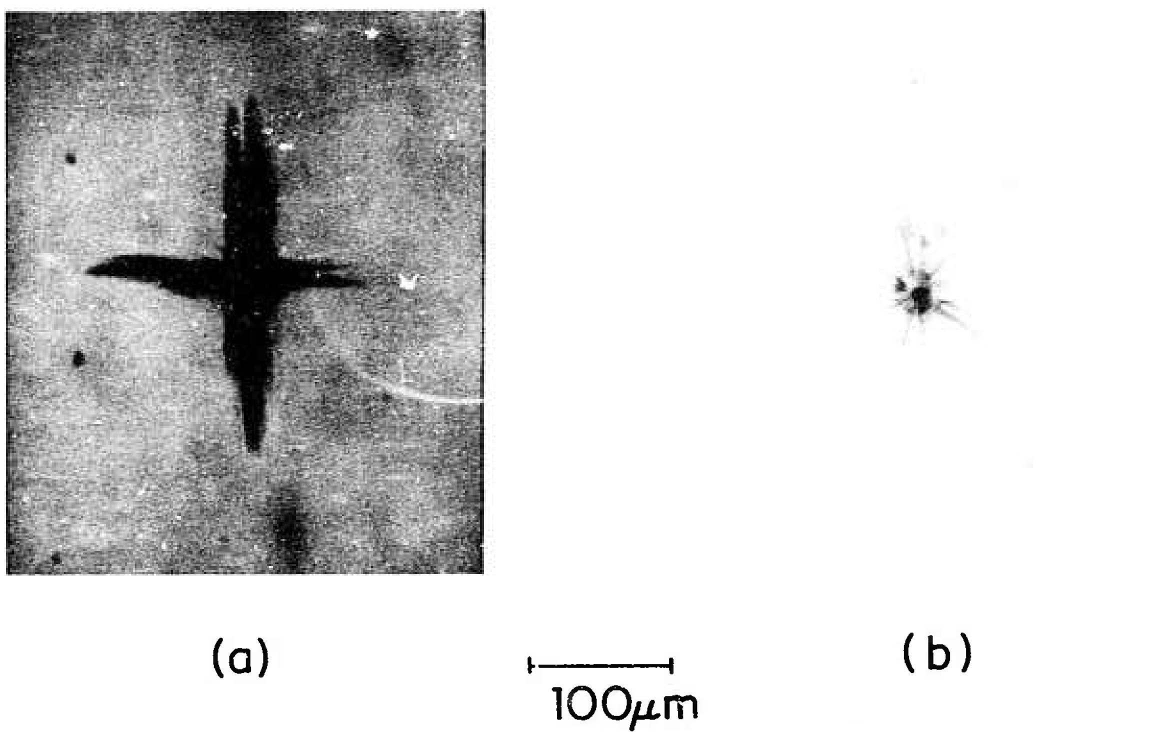


Fig. 5 Stress Induced Fractures in Crystalline and in Glassy $Ba_2MgGe_2O_7$

Residual damage morphology from laser induced damage consists of a small melted region surrounded by stress induced fractures. The direction of light propagation is normal to the plan of the figure.
(a) Fracture in the crystal BMGO occurred along well-defined crystal planes.
(b) In the BMGO glass the directions of fracture were random and different at different damage sites. This behavior is to be expected for an amorphous solid.

System			Damage Intensity*
polycrystal KCl --single-crystal KCl	$E_p / E_{xtal} = 1.0$	$I_p / I = 1.0 \pm 0.02$	$I_p = 8.3$ GW/cm^2
fused quartz -- single-crystal quartz	$E_g / E = 2.2$	$I_g / I = 5 \pm 1$	$I_g = 108$ GW/cm^2
Ba_2MgGeO_3 glass -- single crystal	$E_g / E_{xtal} = 0.9$	$I_g / I_{single\ crystal} = 0.8 \pm 0.2$	$I_{xtal} = 7$ GW/cm^2

* For comparison, the damage intensity for sapphire has been measured in D. Fradin and M. Bass, Appl. Phys. Lett. 22, 157 (1973) to be 110 GW/cm^2 at 1.06 μm with a 10 ns laser pulse. The damage intensity for NaCl, which was used as the standard for the present measurements, is 18 GW/cm^2 as determined in Ref. 10.

TABLE I

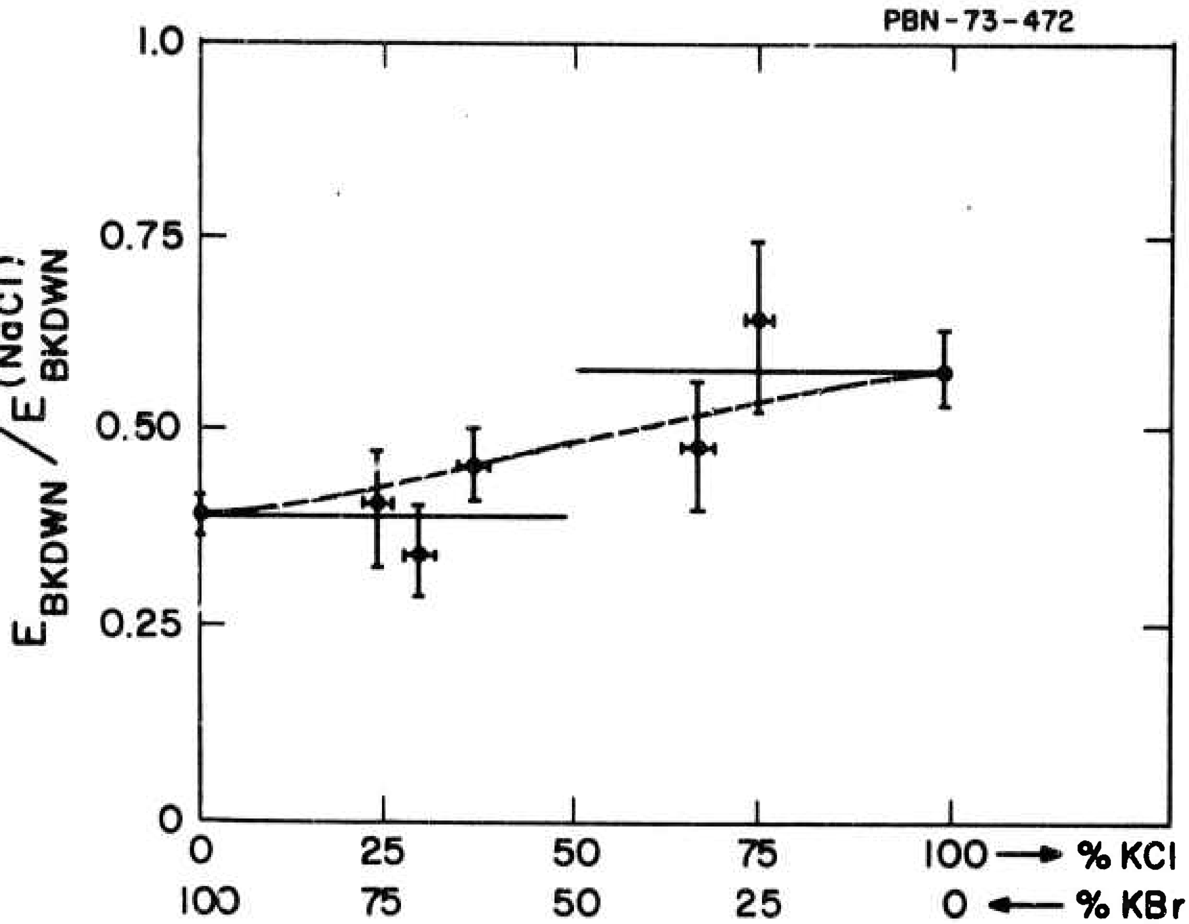


Fig. 6 Intrinsic Breakdown Fields for KBr_xKCl_{1-x} Alloys. The rms breakdown fields are normalized to the breakdown field of NaCl ≈ 2.2×10^6 volts/cm.

greater than the longitudinal optical (LO) energy determine the characteristics of the avalanche. The LO energy in the alkali halides corresponds to electron momenta, k, of about 0.1 times the reciprocal lattice vector, G. Thus the important electrons have $k \gtrsim 0.1G$ and their interaction with the lattice will be dominated by phonons having wave number $\gtrsim 0.1G$. Because such lattice vibrations have wavelengths equal to 10 lattice constants or less, this simple model suggests that the damage field should not be affected by disorder unless the disorder appears on the scale of about 10 lattice constants (~ 50A).[a]

Amorphous systems may be disordered on such a scale. Our observation that fused quartz is more resistant to damage than crystalline quartz is, therefore, consistent with the argument just presented. The data on the $Ba_2MgGe_2O_7$ system is also explained by this argument since that system had regions within the focal volume which were locally ordered on the scale of > > 10 lattice constants. Electron avalanche is apparently initiated in these crystalline regions.

The breakdown fields for various single crystal alloys of KBr and KCl are summarized in Fig. 6. It is seen that the damage strengths of the alloys are intermediate between those of the constituents. There is no evidence that alloying caused disorder sufficient to increase the breakdown strength of these materials. The variation of damage strength with composition can be qualitatively understood by noting that many material parameters such as bandgap, lattice constant, dielectric constant, and phonon frequencies have values intermediate between those of the constituents.[23] According to simple models of avalanche breakdown,[19] the breakdown field depends on these various material parameters so that it is reasonable to expect the breakdown strengths of the alloys to also be intermediate between those of the constituents.

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[a] The larger breakdown field for the amorphous quartz can be understood by another physically equivalent argument. As noted in Refs. 9 and 10, the rate of energy input into the electron population decreases with decreasing electron mobility μ. In low fields μ is smaller in highly disordered systems than it is in crystals with the same chemical composition. If the effective mobility in disordered media is also lower in the ultrahigh fields characteristic of avalanche breakdown, then it should be more difficult to heat the electron distribution in fused quartz, and, as observed, fused quartz should be more damage resistant than crystalline quartz.

It thus appears that extreme lattice disorder such as present in highly amorphous systems has a measurable effect on intrinsic damage fields. This results is qualitatively predicted by simple models of electron avalanche breakdown.

The advice and assistance of D. Bua, R. Newberg, D.W. Howe, C. Willingham and R.R. Monchamp are gratefully acknowledged. A. Linz generously provided several of our samples and J. VanderSande conducted the electron reflection diffraction studies.

IV. DEPENDENCE OF LASER INDUCED BREAKDOWN FIELD STRENGTH ON PULSE DURATION

A. Introduction

Intrinsic damage fields from avalanche breakdowns have been determined for a number of materials using Q-switched laser pulses.[9,10,24] It was found that the damage fields are nearly independent of pulse duration t_p when $t_p \geq 5$ ns. When high frequency ($\geq$ GHz) mode beating occurs, however, the resulting time structure is averaged out by the damage process,[10] thus indicating that characteristic development times of the electron avalanche are of order 10^{-9} sec.

Measurements of avalanche breakdown with subnanosecond laser pulses have not been made prior to the present work. Since laser breakdown and dc dielectric breakdown appear to result from the same fundamental process, data at dc can, in principal, be used to estimate the pulse duration dependence to the laser induced avalanche. Yablonovitch and Bloembergen[25] have, in fact, made such estimates based on measured values of dc breakdown for thin samples of NaCl. Their estimates, however, were based on uncertain limits of an effective, high-field electron drift velocity and ignored important experimental uncertainties such as space charge development which can influence dc breakdown fields but not laser breakdown fields.

There is no reliable data in the literature, therefore, on the laser pulse width dependence of intrinsic optical damage. Such data is useful to our understanding of avalanche breakdown because it provides information on the time development of the electron avalanche. Such data also has practical importance in that it establishes measured upper limits for the propagation of mode locked laser pulses in solids.

We have measured the intrinsic breakdown strength of NaCl at 1.06 μm using four pulse durations between 10.3 ns and 15 ps. It was found that the intrinsic breakdown field increases by a factor of about

6 to over 10^7 V/cm as the pulse duration is changed by about three orders of magnitude. From these measured damage fields, an ionization rate for the electron avalanche can be inferred and compared to the predictions of Yablonovitch and Bloembergen.

B. DEPENDENCE OF LASER INDUCED BREAKDOWN FIELD STRENGTH ON PULSE DURATION [17]

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Abstract

Field strengths at which optical damage is initiated in NaCl have been measured with a mode-locked Nd:YAG laser with pulse durations of 15 and 300 picoseconds. Comparison with previously reported data with a Q-switched laser shows that the field strength required for intrinsic optical damage increases by almost one order of magnitude from 10^6 v/cm at 10^{-8} sec to over 10^7 v/cm at 1.5×10^{-11} sec. This is in qualitative agreement with published estimates based on the electron avalanche breakdown mechanism.

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Damage produced by laser beams of high intensity in transparent materials has been studied intensively for many years. It is only recently, however, that the effects of absorbing inclusions and self-focusing have been carefully eliminated, and the intrinsic breakdown mechanism in transparent condensed dielectric media has been positively identified as electron avalanche ionization.[6-10,18,20] On the basis of this mechanism and known characteristics of dc breakdown by avalanche ionization, Yablonovitch and Bloembergen[25] predicted a characteristic dependence of breakdown field strength on laser pulse duration. In this note an experimental determination of this dependence in the subnanosecond regime is presented, which turns out to be in general agreement with those predictions.

The measurements were performed by focusing mode-locked YAG: Nd laser pulses having durations of 15 and 300 picoseconds inside a single crystal of NaCl. Because the experimental procedures used in the present work were identical to those used in recent studies with a Q-switched YAG: Nd laser,[10] the subnanosecond measurements can be directly compared to the results of these studies. It is found that the intrinsic breakdown field increased by almost an order of magnitude to over 10^7 volts/cm as the laser pulsewidth was decreased from 10 nanoseconds to 15 picoseconds. Previous data with pulse widths typical of Q-switched lasers had demonstrated that the dependence on pulse width in the nanosecond regime is small so that relatively little information on the time dependence of the avalanche process was available.* Our results demonstrate again that lasers can be used to measure properties of dielectric breakdown which are difficult to obtain by dc techniques.[26-28]

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* A pulsewidth dependence to optical damage in thin films was first observed by E.S. Bliss and D. Milam, 4th ASTM Symp. on Damage in Laser Materials, NBS Spec. Pub. 372, 108 (1972).

The mechanism of damage was not identified, however. Since it is known that thin films can have a large residual absorption and significant structural defects and that both effects can change the damage characteristics of surfaces, it is not clear that the change in damage fields which were observed by Bliss and Milam reflect a property of an intrinsic bulk damage mechanism.

The laser used for the present work was a passively mode-locked YAG:Nd laser operating in a TEM_{00} mode at 1.06 μm. Without intercavity etalons, this oscillator produced bandwidth-limited light pulses of 15 picosecond duration. By replacing the output mirror with a sapphire etalon, the pulsewidth was lengthened to about 300 picoseconds. Two-phonon fluorescence measurements failed to detect substructure with pulses of either duration. A laser-triggered spark gap[29] was used to select a single light pulse which, after attenuation, was focused through a 14-mm focal length lens about 2 mm into the sample. Care was taken to insure that spherical aberrations from both the lens and the plane entrance surface of the sample being tested were unimportant. An energy monitor recorded the energy in each laser pulse.

The intrinsic damage process is statistical in nature[18,20] because of the fluctuations in the formation of the first few hot electrons in the small focal volume (about 10^{-7} cm^3). The damage threshold can be defined as that value of the RMS electric field inside the sample which produces damage during one pulse with a probability of 0.5. In NaCl the threshold is quite sharp and damage was identified by the occurrence of a faint spark and concomitant melting of a small region ($\sim 2 \times 10^{9}$ cm^3) inside the crystal. At least 20 data points were taken at each pulse duration at the 0.5 probability point.

Beam distortion from self-focusing was avoided by confining the laser input powers to well below the calculated critical powers for catastrophic self-focusing.[9,10] (See Table II). To verify the absence of self-focusing, two different lenses (focal lengths of 14 and 25 mm) were used to focus the laser radiation. As expected from diffraction effects alone, the input damage powers scaled with the square of the focal lengths. If catastrophic self-focusing had been present with the subnanosecond pulses, the input damage power would have been independent of focal length.[10]

A number of tests have been developed to distinguish between damage from absorbing inclusions and damage from intrinsic breakdown.[9,18] Two of these tests -- examination of the damage morphology and examination of the transmitted laser light -- could not be used in the present work because of the small volumes of the damaged sites and the short durations of the laser pulses. But because breakdown is virtually threshold-like in NaCl, other tests were possible. It was observed that the damage

TABLE II

EXPERIMENTAL BREAKDOWN FIELDS AND CALCULATED SELF-FOCUSING PARAMETERS IN NaCl

Pulsewidth (10^{-12} sec)	P_{input} (10^6 watts)	P_c (10^6 watts)* electrostriction	electronic	E_{rms} (10^6 volts/cm) relative	absolute
15	1.5	2.9×10^4	18	E(15 ps)/E(300 ps)	12.4 ± 3.7
300	0.22	82	18	$= 2.6 \pm 0.7$	4.7
4.7×10^3 †	0.030	1.8	18	E(4.7 ns)/E(10.3 ns)	2.3 ± 0.4
10.3×10^3 †	0.033	1.8	18	$= 1.1 \pm 0.05$	2.1

* P_c is the calculated critical power for catastrophic self-focusing. The calculation of the electrostrictive value is discussed in Ref. 10. The electronic value is estimated from measurements of third harmonic generation.

† Results taken from Ref. 10.

field was well defined and did not change as different regions of the sample were probed and lenses with different focal lengths were used to focus the radiation. Also, only one spark occurred with each damaging laser pulse, and the spark always appeared to form at the geometrical focal plane. These observations contrasted with those obtained under conditions where inclusion damage was seen.[30] It was therefore concluded that except possibly for occasional damage sites, inclusion damage was absent in highly pure NaCl under the conditions of our measurement as it had been in previous work[10] where Q-switched lasers were used.

Table II summarizes the results of the present measurements and those of Ref. 10. An increase in breakdown strength was observed as the duration of the laser pulse was decreased. As the pulse duration was changed from 10.3 nm to 15 ps, there was a total change by a factor of 5.8 in damage field strengths or a factor of 33 in damage intensity. The experimental points are plotted in Fig. 7, together with some semi-empirical predicted curves taken from Ref. 25.

In the subnanosecond regime electron diffusion and self-trapping may be ignored. The density of the conduction electrons in the avalanche changes exponentially with time,

$$N(t) = N_0 \exp[\int \alpha (E)dt] = N_0 M_c(t). \tag{1}$$

When the electron density exceeds about 10^{18} cm^{-3}, requiring a multiplication factor of roughly $M_c \sim 10^8$, breakdown is said to occur. According to Eq. (1) the ionization rate $\alpha(E)_{rms}$ is related to the pulse duration as follows:

$$\alpha (E_{rms}) = t_p^{-1} \ell n\, M_c \approx 18\, t_p^{-1}. \tag{2}$$

This relation has been used to convert the quantity $\alpha(E)$ used along the vertical axis in the figure of Ref. 25 to our figure which uses t_p^{-1}. We have shifted the curves along the horizontal axis to obtain agreement with the experimental values for the breakdown field E_{rms} for the long pulses. It

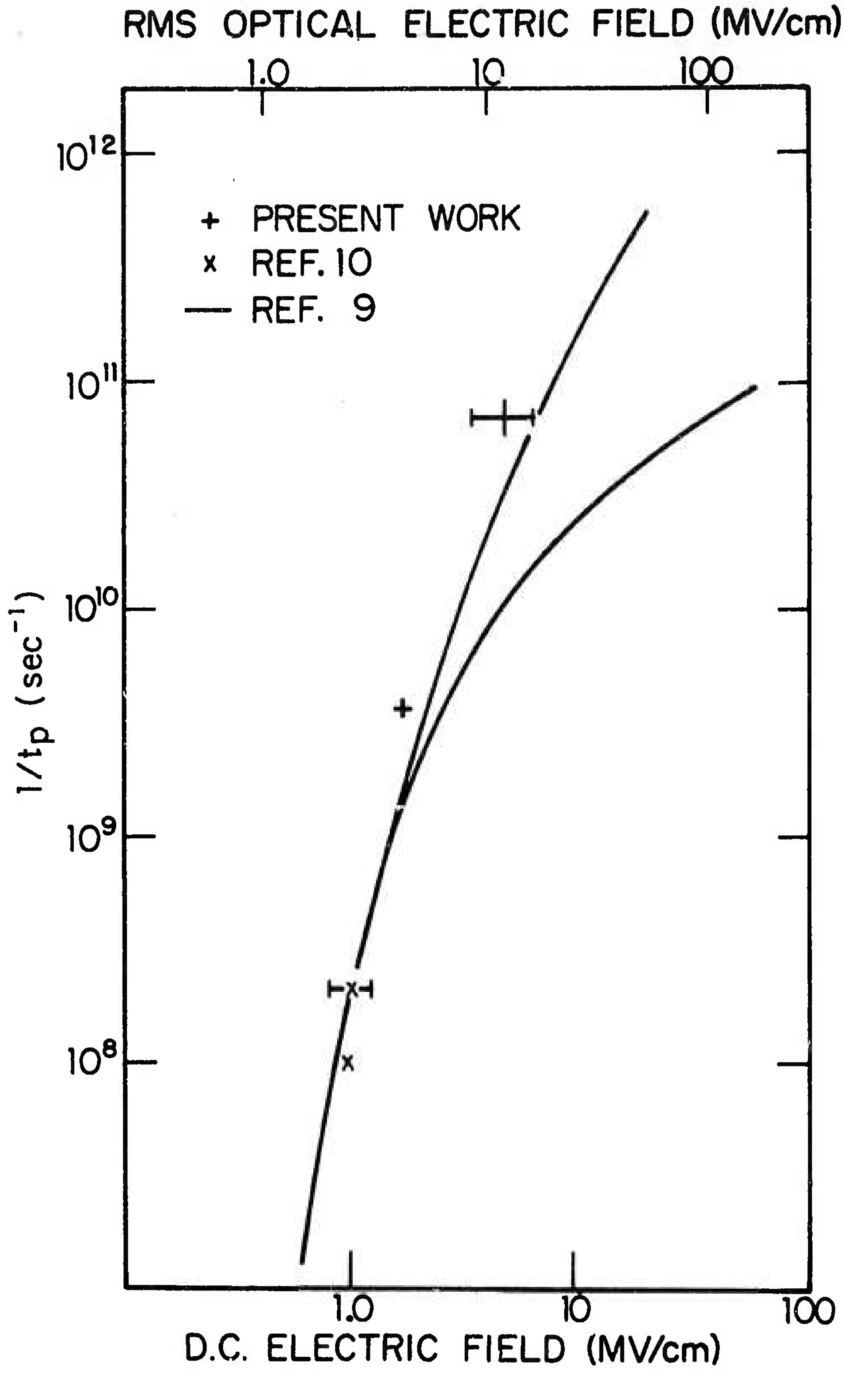

Fig. 7 The Functional Relationship Between the Optical Breakdown Field Strength and the Pulse Duration. The experimental points are compared with two semi-empirical drawn curves. (A discussion of these curves is given in the text and in Ref. 25). The experimental error bars reflect experimental uncertainties in the absolute field strengths.

should be noted that the dc breakdown fields quoted in Refs. 26-28, on which the curves in Ref. 25 were based, are about a factor 2.3 lower than our laser values for E_{rms}. There are reasons to suspect that this factor is due to systematic errors in the dc experiment. First, this factor of 2.3 is nearly the same for all nine alkali halides studied at 10.6 μm[9] and 1.06 μm,[10] and second, dc field values are average values without regard for field inhomogeneities from space charge effects which can be important in dc experiments.[*,31,32] Consistent with the approach of Ref. 9 and 10, it is therefore more meaningful to compare trends in breakdown fields as different materials are investigated or as parameters are varied than to compare the absolute values of breakdown fields.

It is seen that the experimental points fall close to the upper curve of Ref. 25, which was derived on the assumption that the mobility in the hot electron gas is independent of E_{rms}. Quantitative agreement should not be emphasized, however, because the analysis is based on assumptions which are questionable over the range of damage fields considered. As mentioned before, space charge and electrode effect may considerably alter the interpretation of the dc results[19] and consequently to transition from E_{dc} to E_{rms} along the horizontal axis. Furthermore the breakdown field at the shortest pulse durations becomes so high that it is possible that frequency-dependent tunneling (or multiphoton ionization) takes over as an intrinsic damage mechanism. In Ref. 25 it was estimated that this change over would occur for fields higher than 2×10^7 V/cm or pulse durations shorter than one picosecond. If this estimate is inaccurate and this other intrinsic breakdown mechanism begins to compete with avalanche ionization, the trend would be to push the experimental points upward and to the left of the prediction curves based on the avalanche effect alone.

In summary, intrinsic laser-induced damage has been shown to be a time-dependent process. As the laser pulsewidth was decreased to 15 ps, the damage field in NaCl increased to over 10^7 V/cm. From the pulsewidth

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* Prof. Y.R. Shen has suggested (private communication) that local field effects may be important in an electron avalanche and may explain the difference in absolute field strengths between dc and optical frequencies. The damage fields reported in the literature, however, are not corrected for local fields because it is normally assumed that any local field effects are averaged out by the electron's rapid movement across the unit cell. The validity of this latter assumption has not been established.

dependence of the optical damage field, a field-dependent ionization rate was determined and found to agree at least qualitatively with experiments using dc fields. The agreement underscores the basic similarity between intrinsic laser-induced damage at 1.06 μm and dc dielectric breakdown and adds further support to the existence of avalanche ionization in self-focusing "filaments", where effects also occur of a picosecond time scale.

We wish to thank M. Bass, L. Holway, E. Yablonovitch and J.M. McMahon for numerous helpful discussions. M. Bass and J.M. McMahon were particularly helpful in establishing arrangements for this work.

V. THE ROLE OF INCLUSIONS AND LINEAR ABSORPTION IN LASER DAMAGE TO DIELECTRIC MIRRORS

A. Introduction

Recent optical damage studies have established intrinsic limits for light propagation inside transparent solids.[9,10,30] The relationship between surface and bulk damage fields has been determined in previous work by direct comparison of measured intrinsic surface and bulk damage fields[24] and by theoretical analysis.[33] Surface finishing techniques now exist which for selected materials and highly focused laser light, produce effectively imperfection-free surfaces whose damage fields are identical to those of the bulk.[24]

Our understanding of damage to optical coatings, on the other hand, is not nearly as advanced as our understandings of surface and bulk damage. The reason for this difference is that material imperfections such as structural defects, inclusions, and residual absorption have not been sufficiently characterized in dielectric coatings nor have they, in fact, been sufficiently controlled in the manufacturing process.

The present work helps clarify the role of material imperfections in dielectric coating damage. By studying the residual damage morphology and the pulse duration dependence of the damage intensity it is shown that damage thresholds for weakly focused laser lights are determined by highly absorbing inclusions and that there is a large range of inclusion sizes present in the coatings. It is further established that residual absorption in coatings produced from transparent bulk material does not contribute to optical damage.

B. The Role of Inclusions and Linear Absorption in Laser Damage to Dielectric Mirrors

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By studying the morphology of threshold damage and observing the predicted "pulse duration-inclusion size" relationship we have found that laser damage to dielectric coatings is primarily determined by the presence of metallic or highly absorbing nonmetallic inclusions. It is also shown that linear absorption does not determine the damage resistance of coatings when they are properly prepared from materials which do not show bulk absorption.

Key Words: Dielectric mirror, laser-induced damage, inclusions, Linear absorption.

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* Supported in part by the Advanced Research Projects Agency of the Department of Defense and monitored by the Air Force Cambridge Research Laboratories under Contract No. F19628-73-C-0127.

1. Introduction

By studying the morphology of damage produced at threshold and observing the predicted[11,34] "pulse duration-inclusion size" relationship we have found that laser damage to dielectric coatings is primarily determined by the presence of metallic or highly absorbing non-metallic inclusions. Specifically, inclusions less than $\sim 0.4\ \mu m$ in diameter are most easily damaged by single 20 psec duration ruby laser pulses while inclusions with diameters greater than $\sim 3.5\ \mu m$ determine damage resistance due to 20 nsec pulses. This suggests that coating damage resistance to Q-switched pulses can be improved by eliminating the larger inclusions.

To study linear absorption in films, experiments were performed in which the mirror sample was irradiated by a pair of pulses separated by an interval of several nanoseconds. Significant linear absorption would be indicated if neither pulse alone could produce damage, but damage occurred when both pulses were used. These experiments showed that linear absorption in films does not determine the damage resistance of coatings when they are properly prepared from materials which do not show bulk absorption.

2. Experimental Conditions

2.1 Damage Apparatus

Experiments have been performed at a wavelength of 0.69 μm using Gaussian-mode pulses with durations between 20 psec and 20 nsec. The picosecond pulses were selected from a mode-locked pulse train, while the longer pulses were generated in a dye-Q-switched oscillator. For both systems, the temporal profile, pulse energy, and a magnified image of the laser spot incident on the damage specimen are recorded for each firing. Details of the two systems are reported elsewhere.[35,36]

The Q-switched system has recently been modified by the installation of an electro-optic pulse shaper between the oscillator and the amplifier. This has allowed generation of strictly bandwidth-limited pulses 1.4 nsec in duration[37], and pairs of pulses separated by a predetermined and reproducible interval. All damage experiments with shutter-shaped pulses were monitored in the same fashion as has been described elsewhere.[35,36]

2.2 Damage Specimens

With a single exception noted in the next paragraph, all damage specimens were electron-gun deposited, quarter-wave stacks of either TiO_2/SiO_2 or ZrO_2/SiO_2 on fused silica substrates. The reflectivity of each of the mirrors exceeded 90 percent at 0.69 μm. A large number of samples from several manufacturers were examined; the data presented is generally true for all specimens.

A single set of experiments was performed on a ThF_4/ZnS reflector. This exception will be noted in the text.

3. Inclusion Damage in Coatings

3.1 Damage at 20 psec

Characteristic "near-threshold" damage induced by a 20 psec pulse[35,38] focused to a spot 190 μm in diameter (FWHM of the intensity distribution)

is exhibited in the scanning electron micrograph (SEM) of Figure 8. *
The large ridges in the photo on the left are imperfections in the gold coating applied to permit scanning electron microscopy, and are not properties of the dielectric coatings or the laser damage. The laser damage is the array of small craters in the center of this photograph, which corresponds to the central part of the irradiated region. The fact that damage is not produced in the weakly irradiated outer regions is consistent with the assignment of damage threshold to this level of irradiation (3.5 J/cm^2 in 20 psec, or 1.7×10^{11} watts/cm^2).

A magnified view of several of the small craters is shown in the photo on the right. Craters are randomly located in the plane of the coating, but restricted primarily to the top two layers of the stack. There are between 2 and 10×10^6 such craters per cm^2.

Since the size at the top of the craters may be characteristic of explosive rupture of the material, a more meaningful measure of the size of the damage site can be obtained at the bottom of the crater. The mean site diameter obtained using this criterion is 0.2 μm. In no case with a single 20 psec pulse has a site with diameter greater than 0.5 μm been observed.

It should be emphasized that the damage described in this section is caused by a single 20 psec pulse and is not necessarily characteristic of damage produced by a train of several such pulses. The distinction between these two cases will be discussed in Section 4

3.2 Damage at 1.4 nsec

The SEM in Figure 9 illustrates threshold damage caused by a laser pulse 1.4 nsec in duration focused to a diameter of 130 μm at the mirror sample surface. The energy density was 14 J/cm^2. Again we see that the damage is an array of randomly located small craters,

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* Damage sites shown in SEM's used as illustrations in this report are distorted since the specimen is viewed at an angle of 45°. True dimensions are obtained by measurements on a diagonal aligned on the long axis of the ellipse.

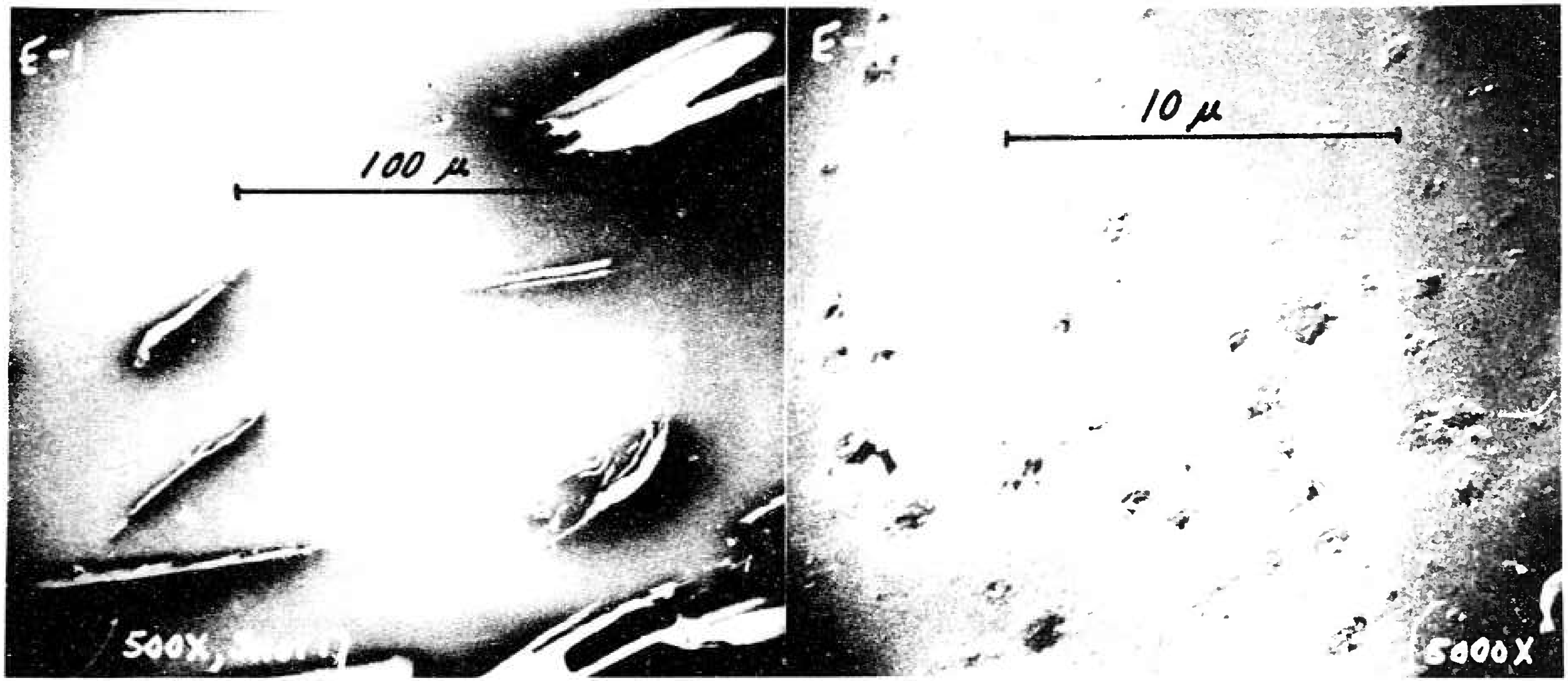

Fig. 8 Laser Damage in a Dielectric Mirror Produced by a Single 20 psec Duration Pulse Focused to a Spot Size of 190 μm (FWHM) in Intensity Profile. Left side: Near-threshold damage consisting of many small craters in the center of the most intensely irradiated area. The large dark features are defects in the gold coating applied to allow electron microscopy. Right side: Magnified view of a portion of the irradiated area.

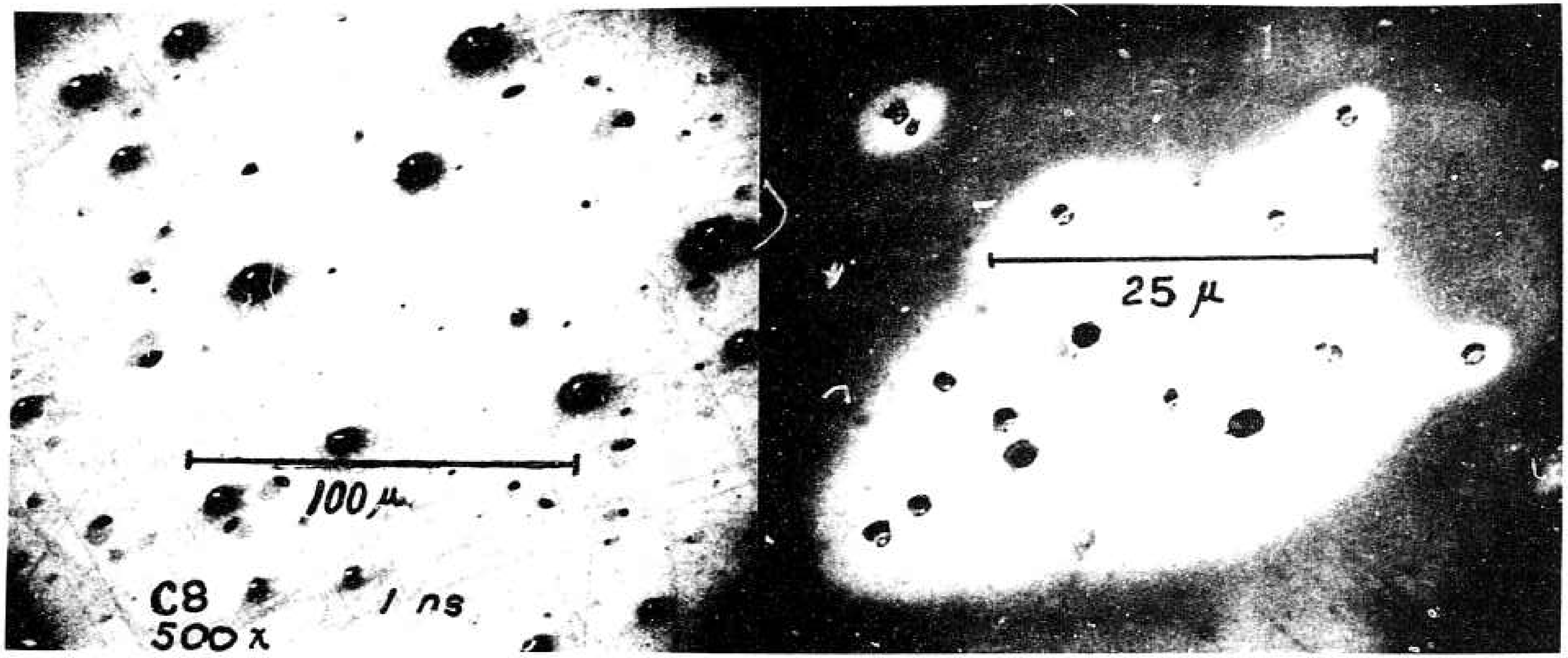

Fig. 9 Laser Damage in a Dielectric Mirror Produced by a Single 1.4 nsec Duration Pulse Focused to a Spot Size of 130 μm (FWHM in the intensity profile). Left side: Near-threshold damage consisting of approximately 20 small craters near the center of the most intensely irradiated area. Dark spots are defects in the gold coating applied to allow scanning electron microscopy. Right side: Magnified view of the damaged region.

only now the mean crater diameter is 1.5 μm. The density of these craters, 10^6 per cm^2, is less than the density of damage sites produced by picosecond pulses. These craters often extend further into the coating than the first one or two layers.

Smaller craters, such as those resulting from irradiation with picosecond pulses, were never produced by these longer laser pulses.

3.3 Damage at 20 nsec

The SEM in Figure 10 illustrates threshold damage caused by a laser pulse 23 nsec in duration focused to a diameter of 400 μm at the sample surface. The energy density was 16 J/cm^2. The mean site diameter was 4.5 μm and the site density was 10^4 per cm^2. In order to measure so low a site density, it was necessary to irradiate a larger area of the sample. Craters produced by 20nsec pulses frequently extend into several layers of the coating.

3.4 Interpretation of Threshold Damage

It has been shown that small impurity inclusions, opaque at the laser frequency and in good thermal contact with the surrounding medium can be heated by a laser pulse to temperatures in excess of the melting point of the medium.[11] The material near such inclusions can undergo a phase change and so laser damage results. When damage due to the presence of inclusions occurs, the residual morphology is characterized by an array of many damage sites randomly located within the irradiated volume.[9, 10, 39] A similar morphology would be expected if the damage were due to any other type of localized material or physical defect.[40]

A morphology characterized by randomly located sites is precisely that of the threshold coating damages shown in Figures 8, 9, and 10. We conclude therefore that localized defects are the principal cause of laser damage to these optical coatings.

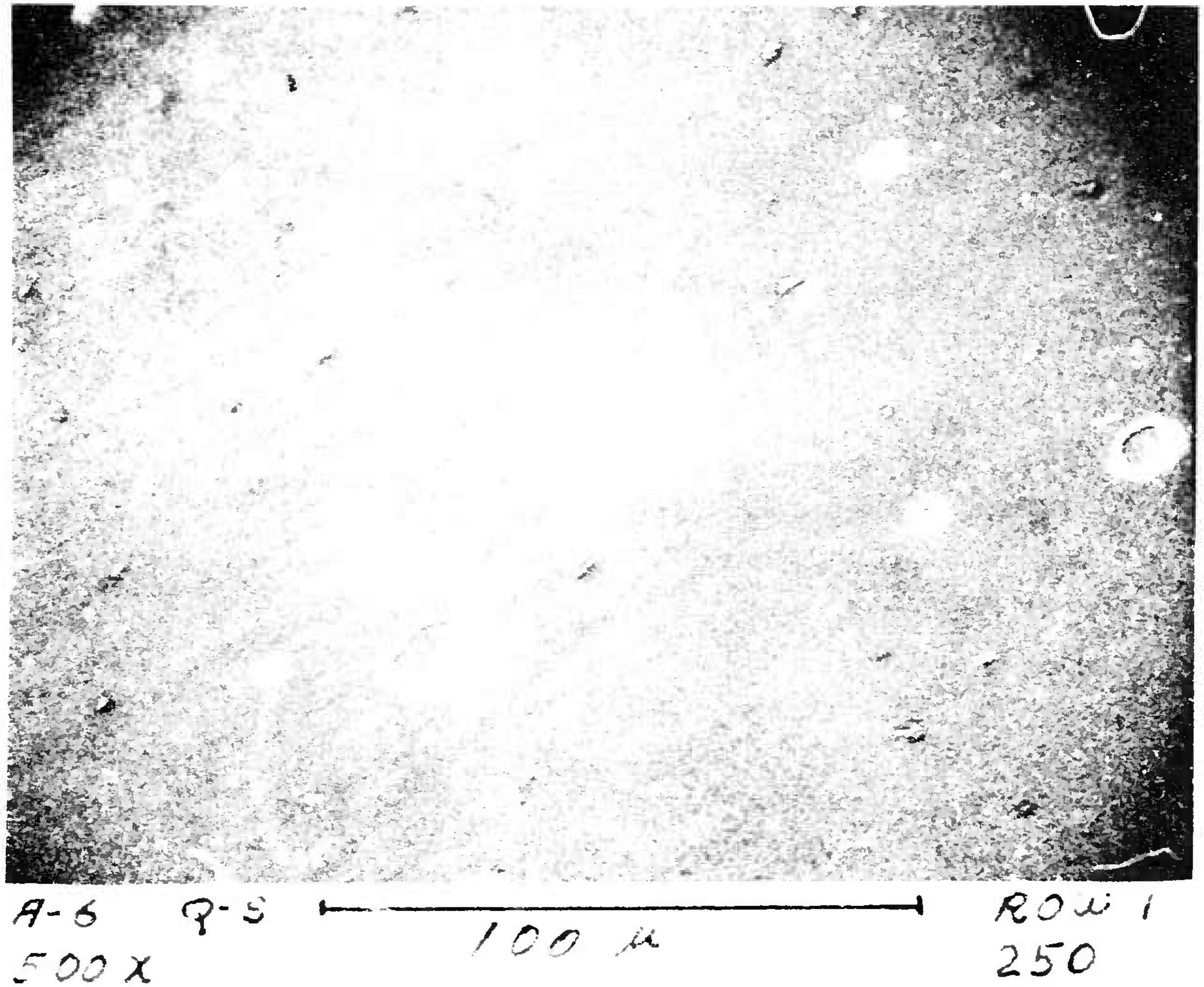

Fig. 10 Near-Threshold Damage in a Dielectric Mirror Produced by 23 nsec Duration Pulse Focused to a Spot Size of 400 μm (FWHM in the intensity profile).

A hint that the defects are impurity inclusions is obtained from the size of the sites damaged by laser pulses of different durations. By considering both the heating and cooling of spherical, metallic inclusions in laser glasses, Hopper and Uhlmann[11] and Bliss[34] have shown that for a particular laser pulse duration a particular size of inclusions will be most easily heated to damaging temperatures.*
These studies show that short pulses can more easily damage small inclusions than long pulses and vice versa. From Eq. 4 in Ref. 11 for the inclusion temperature as a function of the radius, R_i, one can easily show that to a first approximation the inclusion which is most readily heated by a laser pulse of duration τ_p, has radius proportional to $(\tau_p)^{1/2}$. Thus, for the three pulse durations used in this work we would expect the damaged inclusions to have radii in the ratios

$$R_i(23\text{ nsec}):\ R_i(1.4\text{ nsec}):\ R_i(20\text{ psec}) = 34:8.3:1$$

Note that for a particular pulse duration a range of sizes of "easily damaged inclusions" can be expected because the relationship between the temperature reached and the inclusion size is not a very sharply peaked function.[11] In addition, if the level of irradiation is even slightly above the absolute minimum required for damage then several different sizes of inclusions can be heated to damaging temperatures.

Because there is this range of site sizes in each case, the ratios of the mean site diameters,

$$\bar{R}_i(23\text{ nsec}):\ \bar{R}_i(1.4\text{ nsec}):\ \bar{R}_i(20\text{ psec}) = 22:7.5:1$$

are to be compared with the expected ratios. These sets of ratios are in acceptable agreement in view of the difficulty in finding the exact size of the inclusions which produced the damage sites.

Hopper and Uhlmann have also shown that for non-metallic inclusions having moderate absorptivity there is no "pulse duration-inclusion

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* The treatments in Refs. 11 and 34 require that the inclusion be completely embedded in the surrounding material. The morphology of the damage indicates that this condition is satisfied for almost all of the inclusions which contribute to coating damages.

size" relationship as above.[11] However, if the absorptivity of the inclusion is very high (i.e. $1/\alpha < R_i$ where α is the absorption coefficient in cm^{-1} at the laser wavelength) the same model as used for metallic inclusions can be applied. Thus, the observed damage morphology and "pulse duration-inclusion size" relationship suggest that the damaging defects are either metallic or very highly absorbing non-metallic included impurities.

Inclusions of either type may arise from impurities in the starting material, incomplete oxidation of the high index material (particularly in the case of TiO_2), introduction of material from the electron gun,[41] dust, or general deposition of dirt from the chamber. All of these indicate the possibility of improving the damage resistance of optical coatings by improving the coating process control. Note that long pulse damage resistance can be increased simply by eliminating the large inclusions.

Non-metallic absorbing inclusions are likely to be more strongly absorptive at shorter wavelengths and in the infrared than they might be near 1 μm. Thus, their importance in damage problems will grow as the need for high peak power devices in these spectral regions increases.

3.5 Above-Threshold Damage

The morphology of damage sites produced by irradiation at levels significantly above threshold is consistent with the conclusion that damage is caused by metallic or highly absorbing inclusions. In Figure 11 there are four SEM's of damage produced by single 20 psec pulses of successively higher energy. These photographs are highly suggestive that damage for above-threshold irradiation is the result of the compounding of damage at many independent sites. Above-threshold damage with single 1.4 nsec pulses appears to follow the same pattern.

A possible exception to the observation that massive damage is a compounding of damage at many small inclusions occurs with the 20 nsec duration pulses. Above-threshold damage sites at this pulse duration frequently consist of a smooth area, one or two layers deep which is similar

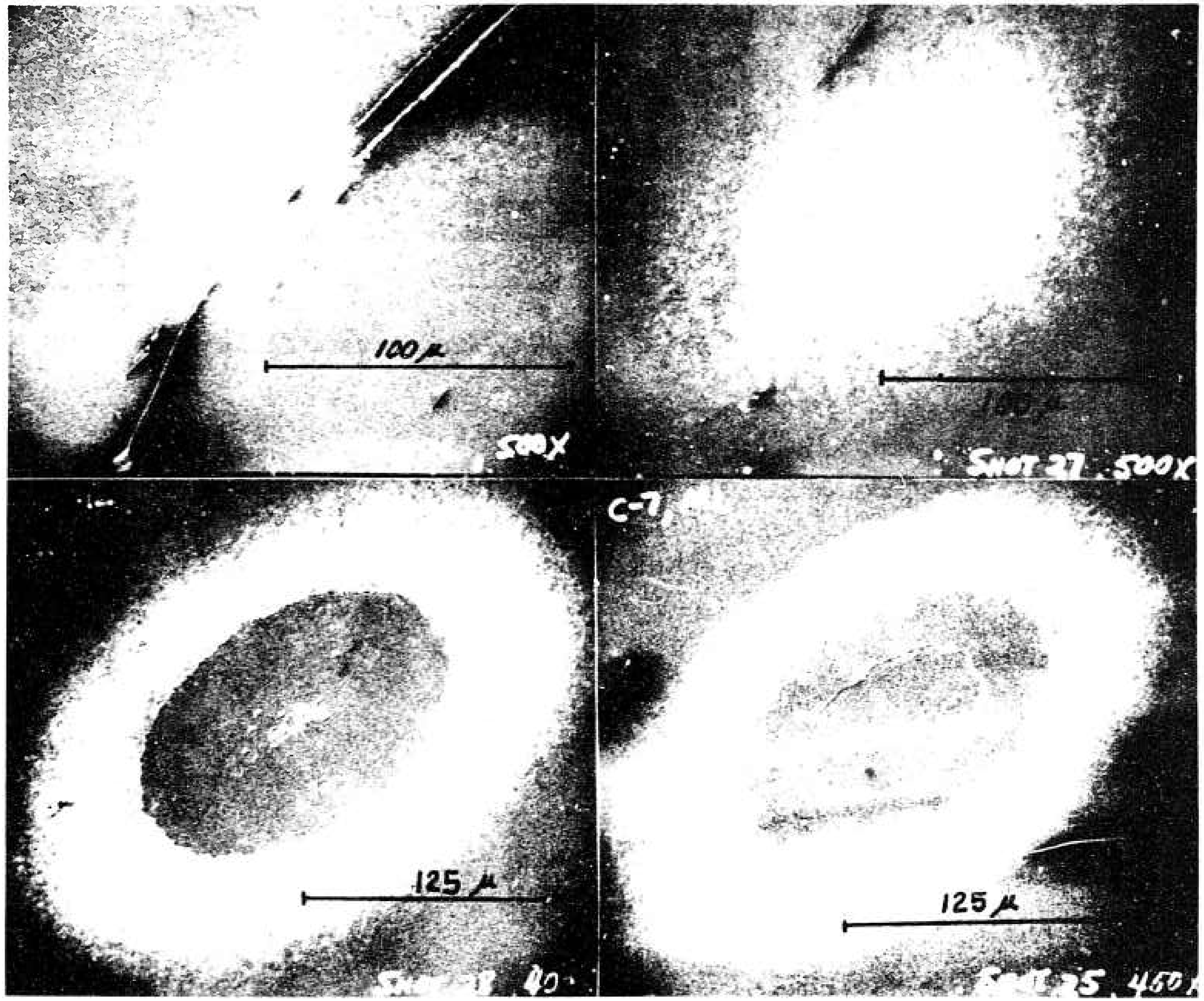

Fig. 11 Damage in a Dielectric Mirror Produced by Single 20 psec Pulses of Successively Higher Energy.

in size and shape to the incident laser beam (see Figure 12). A number of inclusion damage craters may occur either within or outside the edges of the large site. Since the inclusion craters are seldom centered on the larger area, it is not clear that the larger area results from massive inclusion damage. In addition, the material surrounding these sites is not strewn with small inclusion damages as in Figure 11.

The appearance of these large damage sites suggest that other damage interactions might take place with 20 nsec pulses. Numerous mechanisms involving combinations of inclusion damage and/ or intrinsic processes can be invoked to account for the correlation between the shape of the laser beam and that of the larger sites. One possibility is that the outer layers are simply heated to a damaging temperature due to irradiation of the great number of small absorbing inclusions present. Linear absorption by the coating materials will also contribute to a damage morphology similar to the laser beam cross-section. This is explored in detail in the double-pulse experiments described below.

It is to be emphasized that observations concerning the potential importance of other mechanisms for 20 nsec irradiation in excess of threshold in no way effects the conclusion that threshold at this pulse duration is determined by large inclusions.

4. Double-Pulse Damage Experiments

If a damaging quantity of energy, H, is absorbed from a single 1.4 nsec pulse of energy, E, the same energy, H, can be supplied by two pulses each of energy $E_p (E/2 < E_p < E)$ provided that the interpulse interval is shorter than the cooling time of the absorbing volume. Addition of the two pulses to produce damage can occur in a uniformly absorbing film or at inclusions too large to be effectively heated by single 1.4 nsec pulses. This would not occur at small inclusions which could cool during the interpulse interval, or if damage is due to a fast response mechanism such as an electron avalanche breakdown.

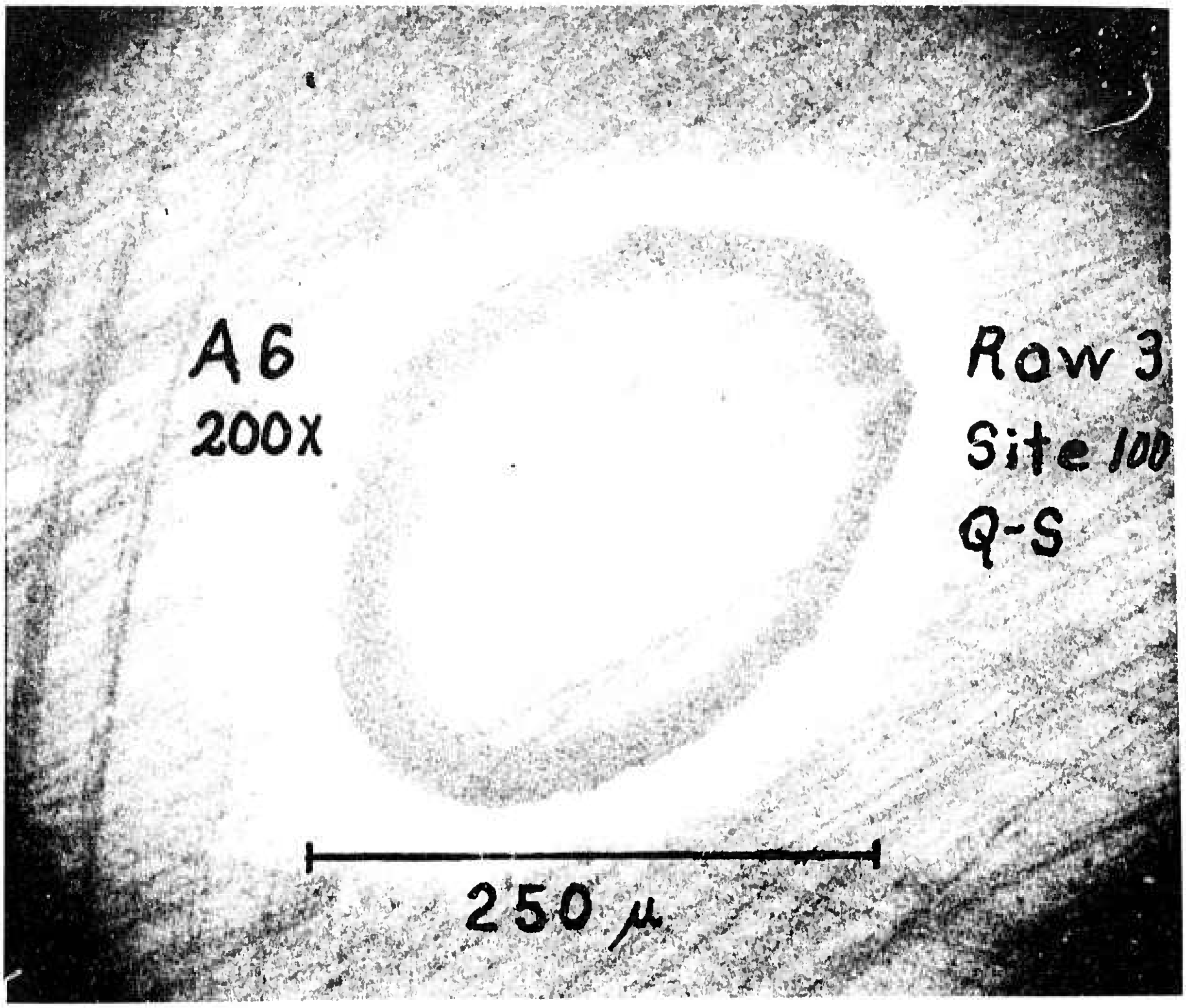

Fig. 12 Above-Threshold Damage in a Dielectric Mirror Produced by a 23 nsec Duration Pulse Focused to a Spot Size of 400 μm. Five small inclusion craters are located within the damaged area which is itself two layers deep. The area has a shape like that of the incident laser beam, and is not centered on the obvious inclusion damage.

As noted in Section 2.1, pairs of optical pulses are readily generated by the pulse-shaping shutter. The Pockels cell in this shutter is constructed as an electrical transmission line element. The driving voltage pulse propagates through the Pockels cell into a cable terminated by an impedance-matched load. Removing the load results in a reflection of the voltage pulse, which in turn opens the shutter a second time. Single-and double-pulse operation of the shutter are both illustrated in Figure 13.

During the experiment, a mirror was probed by single 1.4 nsec pulses at a number of sites to determine the energy E_t, required to produce damage with single pulses. A given site was irradiated only once during this sequence. Additional sites were irradiated with a series of several single subthreshold pulses at intervals greater than one minute. As a general rule, even several pulses with intensities less than that required for single pulse damage produced none. This implies that permanent damage, undetectable by optical microscopy, did not occur with single subthreshold pulses. Subsequent observations of damage due to the addition of two closely spaced pulses could therefore be of thermal origin.

A record of an experiment performed in this fashion is shown in Figure 14. An approximate threshold for damage by single pulses is indicated by the horizontal dashed line, and a lack of cumulative undetected damage by sequences of near-threshold shots which were fired on a given site without causing damage.

A record of some of the double-pulses experiments performed on the same mirror is shown in Figure 15. The total energy ($2E_p$) of each pair of pulses satisfies the requirement $2E_t > 2E_p > E_t$, so that damage was possible only if summation occurred. In several cases, damage did indeed occur. Damage sites produced by addition on this mirror were characteristic of damage due to large inclusions. Such inclusions would not be expected to damage upon irradiation by single 1.4 nsec pulses. This observation, coupled with the failure of addition at many sites is taken as evidence that there is no severe linear absorption present in this mirror beyond that presented by the localized inclusions.

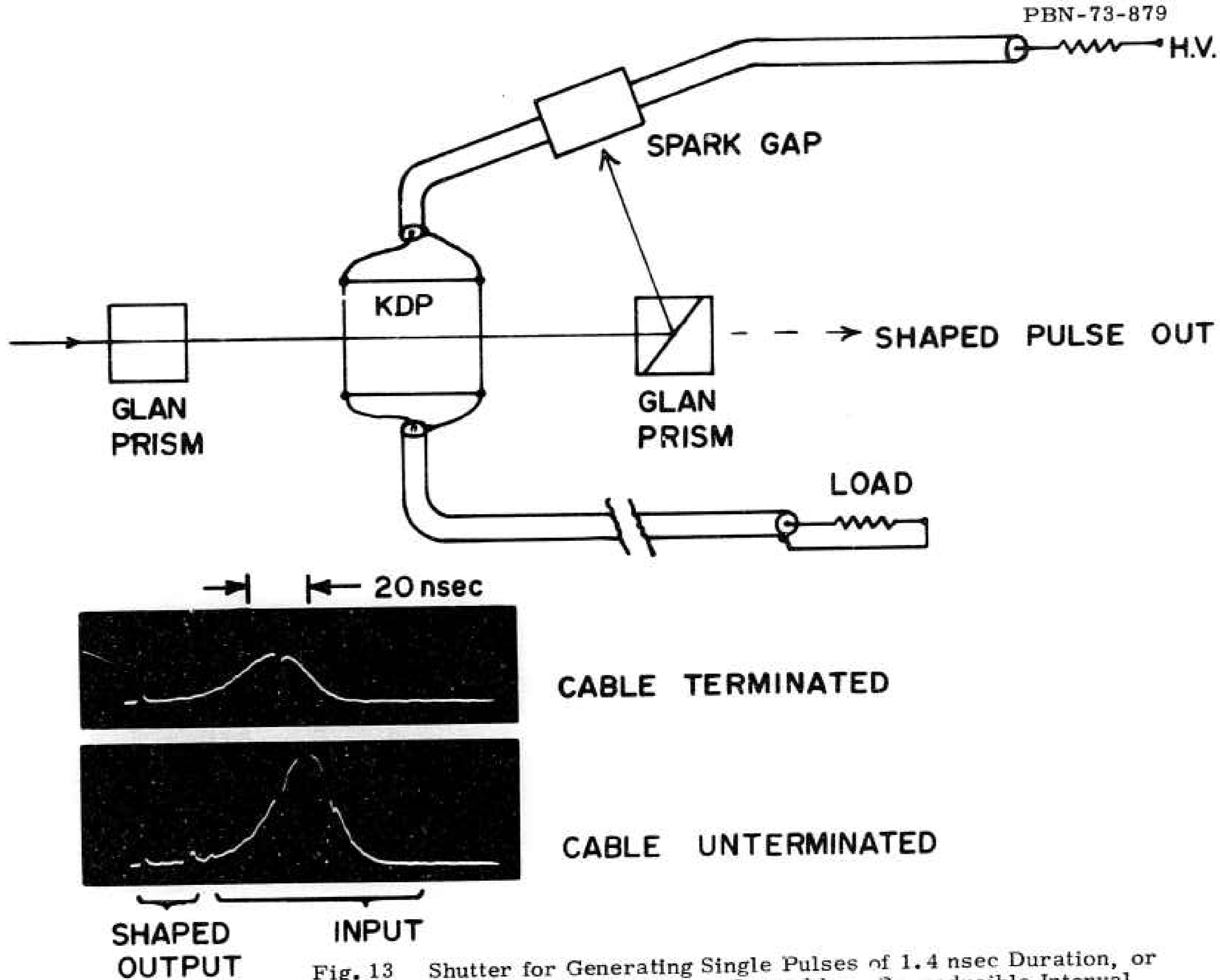

Fig. 13 Shutter for Generating Single Pulses of 1.4 nsec Duration, or Pairs of 1.4 nsec Pulses Spaced by a Reproducible Interval. Removing the load from the cable causes a voltage reflection which gates the Pockels cell a second time.

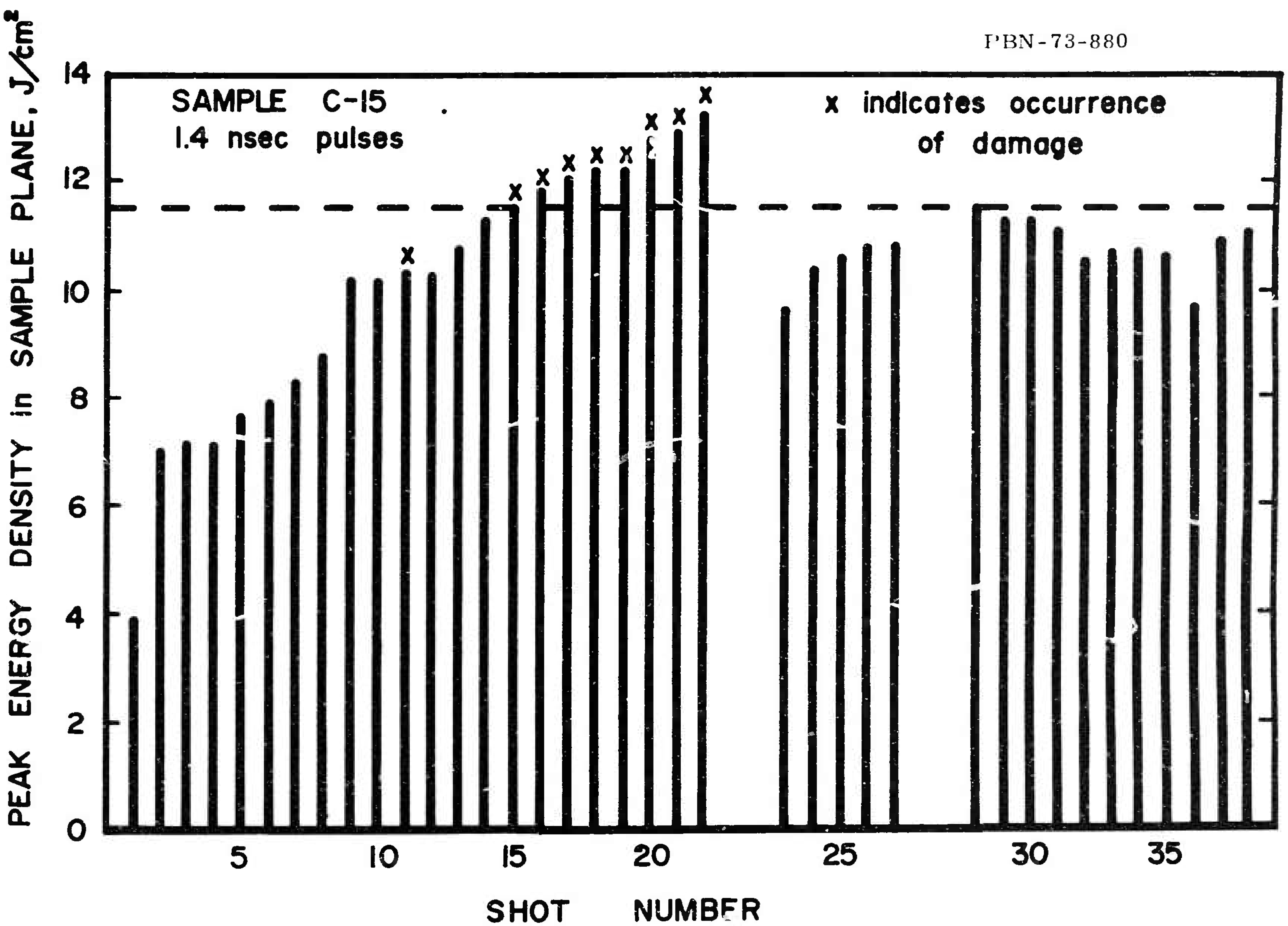

Fig. 14 Histogram of a Damage Experiment on a ZrO_2/SiO_2 Mirror. Shots 1-22 were single shots each fired on a different site to determine single-pulse threshold (indicated by a horizontal dashed line). The set of shots 23-27 were fired onto a single site at one minute intervals to verify that detectable damage did not occur as a result of accumulation of undetected permanent damage. No damage could be detected after the five firings. Shots 28-38 were fired onto a second site at one minute intervals without causing damage.

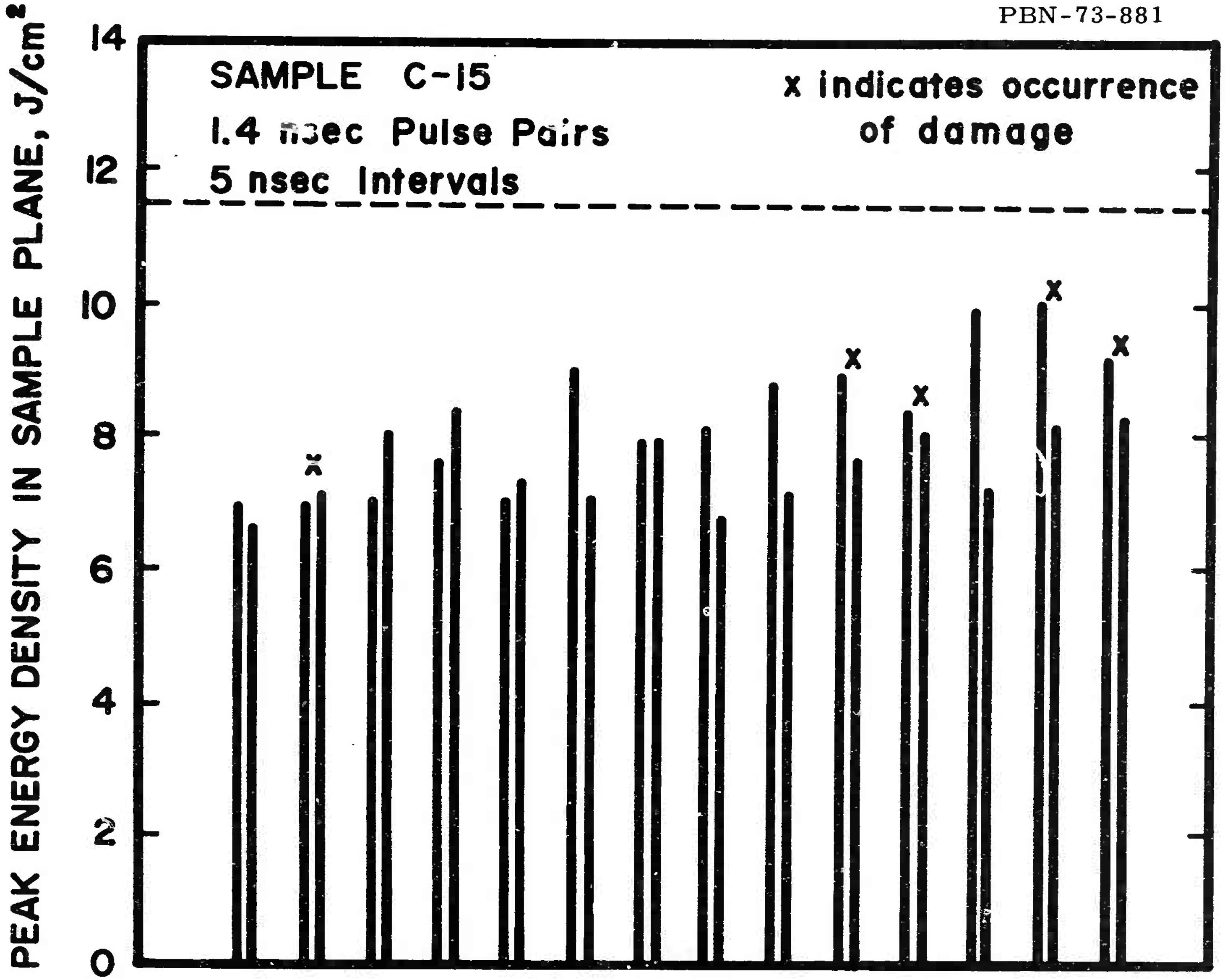

Fig. 15 Record of Some Double-Pulse Experiments on the Sample for Which Single-Pulse Data is Shown in Fig. 16. The failure to observe a consistent pattern of damage due to pulse-pair summation indicates that linear absorption in the film itself is not a limiting problem. A study of the morphology of the sites at which damage did occur by summation shows that damage was due to a class of larger inclusions than would have been damaged by single 1.4 nsec duration pulses.

The observation that large inclusions can be damaged due to cumulative heating by successive pulses can be generalized to explain damage by a train of mode-locked pulses. If the intensity of each of the pulses is such that singly they cannot produce damage, damage to large inclusions can still be the principal failure mechanism. This may explain the difference between the damage threshold found for single 20 psec pulses, $\sim 10^{11}$ watts/cm^2, and the intensity limits of mode-locked oscillators set by coating damage, $\sim 1\text{-}4 \times 10^9$ watts/cm^2.

It was a general observation that there was no consistent pattern of pulse-pair addition indicative of material absorption in the mirrors used in this study.

In order to ascertain that the double-pulse experiment does indeed detect absorption, we have used the technique to study a ThF_4/ ZnS reflector which is known to be linearly absorptive at $\lambda = 0.69\ \mu m$. Typical double-pulse data for this mirror is shown in Figure 16, where the single-pulse damage threshold is again represented by the dashed line. For this reflector, the expected consistent addition of pulses is seen to occur.

These results indicate that if the coating materials are non-absorbing than a combination of linear absorption and/or absorption by small inclusions cannot be invoked to explain results such as those shown in Figure 12. It is possible therefore that this type of damage was caused by an intrinsic process. To study this possibility, experiments in which a spot size sufficiently small such that the irradiated area of the coating is not likely to contain inclusions which are easily damaged by the pulses used are in progress.

5. Conclusions

The principal conclusions of this work are:

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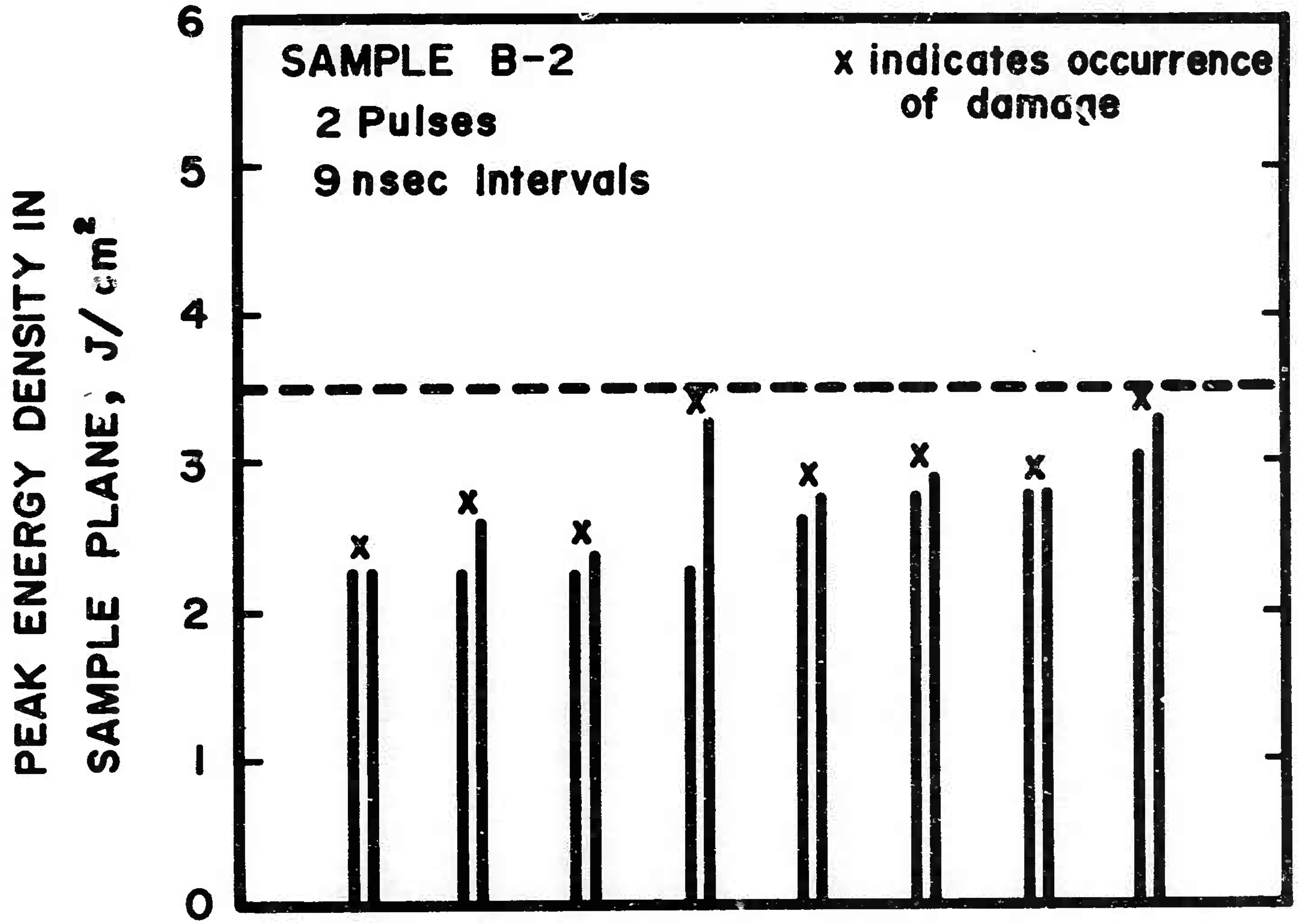

Fig. 16 Double-Pulse Damage Experiments on a ThF_4/ZnS Mirror. Single-pulse threshold is indicated by the dashed horizontal line. A consistent pattern of damage due to addition of two subthreshold pulses indicates linear absorption in the coating.

1. The practical damage threshold for optical coatings at 0.69 μm is determined by the presence of absorbing impurity inclusions.

2. The predicted "pulse duration-inclusion size" relationship for metallic or highly absorbing non-metallic inclusions was observed.

3. Very small (< 0.3 μm) inclusions are the limiting factor in coating damage resistance to single 20 psec ruby laser pulses.

4. Larger inclusions (> 4 μm) are responsible for threshold coating damage when long duration pulses (≈ 20 nsec) or trains of mode-locked pulses are used. Damage resistance to such irradiation would be improved if only this class of larger inclusions could be eliminated. Inclusions of these sizes are appropriate to laser radiation in the visible or near infrared. For other wavelengths the limiting inclusion sizes must be scaled according to the wavelength.

5. Linear absorption in films does not determine the damage resistance of coatings when they are properly prepared from materials which do not show bulk absorption.

6. Acknowledgments

The authors wish to acknowledge the use of data collected by E. S. Bliss (currently with Lawrence Livermore Laboratories) while he was employed at Air Force Cambridge Research Laboratories. They also acknowledge the assistance of C. C. Gallagher in design and construction of the laser-triggered spark gaps and fast pulse circuitry, and the assistance of E. E. Hoell and H. Miller in collecting some of the data presented here. The latter three individuals are employed at Air Force Cambridge Research Laboratories.

VI. PLANS FOR THE REMAINDER OF THE PROGRAM

A. CO_2 Laser

The design and construction of an operating CO_2 laser is complete. Before the laser can be used for damage measurements, however, it will be necessary to design the cavity optics to produce TEM_{00} output with at least 150 KW of peak laser power. Because the TEM_{00} mode for a simple cavity fills only a small fraction of the available resonator volume, more sophisticated optical designs, as the TRIOPTIC[12] or unstable resonator configuration,[13] may be required to reach our specifications. Longitudinal mode selection will be attempted and possibly incorporated into the final cavity design.

B. Damage to ZnSe

Because of its great potential importance as an infrared laser material, damage to ZnSe will be studied during the final phase of the program. Measurements will be made at 1.06 μm and 10.6 μm. In addition, the size and distribution of damaging material defects will be characterized by varying the degree of external focusing.

C. Other Work

The relationship between self-focusing and optical damage will be studied analytically by use of the paraxial ray approximation and estimates of self-focusing parameters will be obtained from experimental techniques based on this analysis. Measurements of the intrinsic breakdown strengths of YAG and YAlO will be made at 1.06 μm.

VII. REFERENCES

1. P.R. Pearson and H.M. Lamberton, IEEE J. Quant. Elec. QE-8, 145 (1972).

2. T.Y. Chang and O.R. Wood, IEEE J. Quant. Elect. QE-8, 721 (1972).

3. A.J. Bealieu, Appl. Phys. Lett. 16, 504 (1970).

4. A.J. Bealieu, Proc. IEEE 59, 667 (1971).

5. R. Dumanchin, Post deadline paper 13 A-6, IEEE, Laser Eng. and Appl. Conf. Washington (1971).

6. A.K. Laplamme, Rev. Sci. Instr. 41, 1578 (1970).

7. J.D. Cobine, Gaseous Conductors (McGraw-Hill, New York, 1941), p. 177.

8. D. Bua and D. Wilson, unpublished.

9. E. Yablonovitch, Appl. Phys. Lett. 19, 495 (1971).

10. D.W. Fradin, E. Yablonovitch and M. Bass, Appl. Optics 12, 700 (1973).

11. R.W. Hopper and D.R. Uhlman, J. Appl. Phys. 41, 4023 (1970).

12. D.R. Whitehouse, C.F. Luck, C.P. VonMertons, F.A. Horrigan, and M. Bass, Paper A-7, IEEE, Laser Eng. and Appl. Conf., Washington (1973).

13. A.E. Siegman and R. Arrathoon, IEEE J. Quant. Elect. QE-3, 156, (1967).

14. D. Bua, D. Fradin and M. Bass, IEEE J. Quant. Elect. QE-8, 916, (1972).

15. J.A. Weiss and L.S. Goldberg, IEEE J. Quant. Elect. QE-8, 758, (1972).

16. A. von Hippel and R.J. Mauer, Phys. Rev. 59, 820 (1941).

17. D. Fradin, N. Bloembergen and J.P. Letellier, Appl. Phys. Lett. 22, 635 (1973).

18. M. Bass and D. Fradin, IEEE J. Quant. Elect., QE-9, 890 (1973).

19. J.J. O'Dwyer, The Theory of Dielectric Breakdown of Solids, (Oxford University Press, London, 1964).

20. M. Bass and H.H. Barrett, IEEE J. Quant. Elect. QE-8, 338 (1971).

21. M. Munasinghe and A. Linz, Phys. Rev. B4, 3833 (1971).

22. J. VanderSande, MIT, private communication (1973).

23. J.H. Fertel and C.H. Perry, Phys. Rev. 184, 874 (1969).

24. D.W. Fradin and M. Bass, Appl. Phys. Lett. 22, 157 (1973).

25. E. Yablonovitch and N. Bloembergen, Phys. Rev. Lett. 29, 907 (1972).

26. A.A. Vorob'ev, G.A. Vorob'ev, and L.T. Musashko, Fiz. Tverd. Tela 4, 1967 (1962) [Sov. Phys. Solid State 4, 1441 (1963)].

27. D.B. Watson, W. Heyes, K.C. Kao, and J.H. Calderwood, IEEE Trans. Elec. Insul. 1, 30 (1965).

28. G.A. Vorob'ev, N.I. Lebedeva, and G.S. Nadorova, Fiz. Tverd. Tela 13, 890 (1971) [Sov. Phys. Solid State 13, 736 (1971)].

29. J.P. Letellier, Naval Research Laboratory Report 7463 (1972).

30. D.W. Fradin and M. Bass, Appl. Phys. Lett. 22, 206 (1972).

31. H. Raether, Electron Avalanches and Breakdown in Gases (Butterworths, London, 1964).

32. J.R. Hanscomb, J. Appl. Phys. 41, 3597 (1970).

33. N. Bloembergen, Appl. Optics 12, 661 (1973).

34. Bliss, E.S., "Opto-Electronics 3, 99-108 (1971).

35. Bliss, E.S. and Milam, D., "Laser Induced Damage to Mirrors at Two Pulse Durations," Proc. 4th ASTM Symp. Damage in Laser Materials, NBS Spec. Pub. No. 372 (1972).

36. Bliss, E.S. and Milam, D., "Laser Damage Study with Subnanosecond Pulses," AFCRL Report No. 72-0233 (1972). Available from Defense Documentation Center, the National Technical Information Center, or the authors.

37. Milam, D., Bradbury, R.A., and Gallagher, C.C., "Evaluation of Three Techniques for Producing Laser Pulses of Nanosecond Duration," AFCRL Report No. 73-0007. Available from Defense Documentation Center, the National Technical Information Center, or the authors.

38. Bliss, E.S., Milam, D., and Bradbury, R.A., Applied Optics 12, 602 (1973).

39. Bass, Michael, IEEE J. Quant. Elect. QE-7, 350 (July 1971).

40. DeShazer, L.G., "Role of Coating Defects in Laser Induced Damage to Thin Films," Proc. 5th ASTM/NBS Boulder Damage Symposium, to be published.

41. Schwartz, H., "Thin Films of Metals and Inorganic Compounds Vacuum Deposited by High Energy Laser" in "Laser Interactions and Related Plasma Phenomena", Vol. I, Ed. by Helmit J. Schwarz and Heinrich Hora (Plenum Press, New York, N.Y. 1972) p. 71.

APPENDIX A

Abstract of Talk Presented by D.W. Fradin at the
Winter Meeting of the American Physical Society
New York City
January 29 - February 1, 1973

Electron Avalanche Breakdown at Optical Frequencies.* D.W. Fradin, Harvard University, and M. Bass, Raytheon Research Division. -- Recent measurements of intrinsic laser-induced damage in alkali halides have been made at 10.6 and 1.06 μm.[1,2] These studies have shown that the damage mechanism is an electron avalanche which is fundamentally the same as dc dielectric breakdown. Experiments have now been performed at 0.69 μm with a ruby laser, and the effects of frequency dispersion in the avalanche are seen, indicating that at this high frequency the process is no longer in its dc limit. In addition, preliminary measurements of the pulse-width dependence to the damage threshold have been made. From these results it is possible to infer an approximate electron-phonon collision frequency appropriate to fields of the order of 10^6 volts/cm, the temporal behavior of the electron avalanche, and an estimate of a high-field electron mobility.

* Supported by Joint Services Electronics Program and by the Advanced Research Projects Agency.

[1] E. Yablonovitch, Appl. Phys. Lett. 19, 495 (1971).

[2] E.W. Fradin, E. Yablonovitch, and M. Bass, to be published, Applied Optics (April 1973).

Abstract published in Bull. Am. Phys. Soc. II 18, 74, (1973).

APPENDIX B

Abstract of Talks Presented by J.P. Letellier and D.W. Fradin at the
Twelfth Symposium on Electron, Ion, and Laser Technology
Massachusetts Institute of Technology
Cambridge, Massachusetts
May 21 - 23, 1973

Dependence of Laser Induced Breakdown Field Strength on Pulse Duration

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Damage produced by laser beams of high intensity in transparent materials has been studied intensively for many years. It is only recently, however, that the effects of absorbing inclusions and self-focusing have been eliminated, and the intrinsic breakdown mechanism in transparent condensed dielectric media has been positively identified as electron avalanche ionization.

Measurements have now been made of intrinsic optical damage induced by subnanosecond laser pulses. These measurements were performed by focusing single, TEM_{00} mode-locked laser pulses having time durations of 15 and 300 picoseconds inside single crystal NaCl. Because the experimental procedures used in the present work were identical to those used in previous studies with a Q-switched YAG:Nd laser, the subnanosecond measurements can be directly compared to the results of those studies. It was found that the root-mean-square intrinsic breakdown field increased by almost an order of magnitude to over 10^7 volts/cm as the laser pulsewidth was decreased from 10 ns to 15 ps.

The dependence of the damage field on laser pulse duration is used to calculate a field-dependent ionization rate which is compared to the predictions of Yablonovitch and Bloembergen who estimated the ionization rate from published measurements of the dc dielectric strength of NaCl for thin samples with varying thickness. Qualitative agreement is found. Such agreement underscores the basic similarity between intrinsic laser-induced damage at 1.06 μm and dc dielectric breakdown and add further support to the existence of avalanche ionization in self-focused "filaments" where effects also occur on a picosecond time scale.

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† Supported by NRL Problem K03-08.502, ARPA Project Order 2062, Project 62301D.

Effects of Lattice Disorder on the Intrinsic Optical Breakdown Strength of Transparent Solids

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Measurements will be reported which probe the effects of material disorder on the intrinsic optical bulk breakdown fields of transparent solids. Complications from self-focusing and inclusions have been carefully avoided. Three disordered materials were studied using a single-mode, Q-switched YAG:Nd laser -- polycrystal KCl, a single crystal KBr-KCl (67% - 33%) alloy, and fused quartz. In each case the damage field for the disordered material is compared to the optical strength of the corresponding single crystal. These measurements were made in order to determine if the optical breakdown field increases with severe lattice disorder as has been observed in dc breakdown experiments and as predicted by simple theories of avalanche breakdown.

It was found that the damage field of the large-grain (20 μm) polycrystal was the same as that measured in the single crystal and that the alloy damage field was intermediate between the breakdown fields of the pure constituents, being about 20 percent larger than the damage field measured in KBr. In the quartz system, on the other hand, the disordered (amorphous) phase was noticeably stronger than the crystal, the ratio of damage intensities being 5 ± 1. This ratio is identical to the corresponding ratio of surface damage fields measured by Bass and Barrett.

Arguments will be presented which explain these results in terms of electron avalanche breakdown.

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The Relationship Between Self-Focusing and Optical Bulk Damage and the Measurement of Self-Focusing Parameters*

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When catastrophic self-focusing occurs in solids, the results are dramatic -- an extended spark and local disruption of the crystal. Because such optical damage apparently always accompanies catastrophic self-focusing in crystals, it has been customary in the damage literature to associate damage with collapse of the laser beam. Results of recent experiments show, however, that it is possible to induce optical damage without measurable beam distortion from self-focusing. This is possible because catastrophic self-focusing and beam distortion depend on the laser incident power in the steady-state whereas intrinsic optical damage depends on intensity for a fixed laser pulse duration.

The relationship between optical damage and self-focusing will be discussed within the framework of a paraxial ray approximation. It will be shown that optical damage can be studied without the occurrence of catastrophic self-focusing and that beam distortion can be minimized by confining laser input powers to values at least an order of magnitude below calculated critical powers for self-focusing while using strong external optics to achieve the high intensities necessary to induce damage. Experimental tests will be summarized which are useful for confirming the absence of catastrophic self-focusing and establishing limits on beam distortion.

A new technique will be described which uses results of the paraxial ray analysis and experimental measurements of optical damage to determine values for the nonlinear index n_2. This technique consists of determining the powers necessary to induce intrinsic bulk damage as lenses with different focal lengths are used to focus the radiation and then comparing these results to the analytical calculations. High input powers are not needed so that relatively small, stable lasers can be used. As an illustration of the application of this technique, the self-focusing parameters for sapphire will be determined.

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* Work supported by the Joint Services Electronics Program at Harvard University under Contract No. N00014-67-A-0298-0006 and the Advanced Research Projects Agency of the Dept. of Defense at Raytheon Research Division as monitored by the Air Force Cambridge Laboratories under Contract No. F19628-73-C-0127.

APPENDIX C

Paper Presented by D.W. Fradin at the
5th NBS-ASTM Laser Damage Symposium
Boulder, Colorado
May 1973

Studies of Intrinsic Optical Breakdown

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Previous work demonstrated that intrinsic optical damage is caused by electron avalanche breakdown. We have conducted a number of recent studies of intrinsic damage which have reinforced the original identification of the damage process and which have probed various characteristics of avalanche breakdown.

By using a ruby laser to induce damage in the alkali halides, we have observed frequency dispersion in the relative breakdown fields. This dispersion, which was not apparent at 1.06 μm, provides insight into the development of the avalanche. A mode-locked Nd:YAG laser with output pulses of 300 and 15 ps duration was used to induce damage in NaCl, and the results were compared to Q-switched studies. It was found that the rms breakdown field increased by almost an order of magnitude to over 10^7 volts/cm as the pulse duration was reduced from 10 ns to 15 ps. This result agrees at least qualitatively with published dc breakdown measurements. A statistical character to bulk optical damage was observed in a number of materials including sapphire and fused quartz and found to be indistinguishable from statistics observed in surface damage. This observation and measurements of the ratio of surface to bulk breakdown fields show that the intrinsic mechanisms for surface and bulk breakdown are identical. Finally, the effect of crystalline disorder on the breakdown strength of solids was studied by measuring the intrinsic damage fields for a polycrystal, an alloy, and an amorphous insulator and comparing the results to the damage fields for the respective single crystals.

1. Introduction

It was determined in previous work [1,2] that intrinsic optical damage in transparent solids occurs as the result of an electron avalanche. During the past year we have extended the original studies of intrinsic breakdown in solids in order to ascertain the dependences of intrinsic damage fields on laser and material characteristics.

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† Supported by the Advanced Research Projects Agency of the Department of Defense and was monitored by the Air Force Cambridge Research Laboratories under Contracts No. F19628-70-0223 and No. F19628-73-C-0127.

Our recent work was conducted for two reasons. The first was to gather experimental information which could provide a basis for a realistic theory of avalanche breakdown. Although avalanche breakdown has been studied for over 40 years [3], very little progress has been made in understanding the details of an avalanche process. Not only is the proper modeling of this highly complex process beyond the present level of solid state theory, [4] but experiments on dc breakdown are difficult to conduct and often hard to interpret. [3] For example, no estimates of effective electron collision rates can be obtained from dc experiments, and the time dependence of the avalanche can only be studied indirectly. [5] Avalanche breakdown at optical frequencies, on the other hand, is experimentally simpler to study and the great versatility of the laser can be exploited to probe aspects of the process that are inaccessible to dc investigators.

The second reason for conducting this work was to answer the following important practical questions: (1) What are the magnitudes of intrinsic damage fields in various materials? (2) How do these fields change with increasing laser frequency and decreasing pulse duration? (3) What is the relationship between surface and bulk optical damage? This information is important to the development of more damage resistant surfaces and coatings and to establish measured upper limits for material damage resistance.

In Sects. 2 and 3 new and previously published results are presented which extend the damage measurements in the alkali halides to higher frequencies and shorter pulse durations. It will be shown, in particular, that frequency dispersion begins to develop at 0.69 μ m and that the damage field in NaCl increases with decreasing pulse width. These two observations provide an experimental estimate of the high-field electron-phonon collision frequency and an experimental measure of the avalanche ionization rate. An apparent anomaly in the frequency dispersion suggests the possibility that deep lying exciton levels may affect the breakdown strength of NaF. A comparison of the optical frequency ionization rate to estimates based on dc data [5] show at least qualitative agreement. A short experiment described in Sect. 4 investigates the effects of lattice disorder on the intrinsic breakdown strengths of materials.

In Sect. 5 data will be summarized which show that the bulk damage process in a number of materials, including NaF, has a statistical character. Measurements of the distribution of breakdown starting times are compared to the surface damage experiments of Bass and Barrett [6] and found to be consistent with those experiments. Sect. 6 summarizes a study which confirms Bloembergen's model of electric field enhancement at polishing defects in optical surfaces. This enhancement makes conventionally finished surfaces damage more easily than the bulk while imperfection free surfaces break down at the same field which is required to produce internal breakdown. Finally, some of the implications of our experimental results for avalanche theory are discussed in Sect. 7.

2. Avalanche Breakdown Induced by Ruby Laser Light

We have completed measurements of intrinsic bulk breakdown in nine single crystal alkali halides using a TEM_{00}, single-longitudinal mode ruby laser. Since this work has already been published, only the result will be given here. [8] Self-focusing was absent in these studies, and damage from inclusion absorption was distinguished from intrinsic damage. It was found that at 0.69 μm the relative breakdown strengths of the alkali halides have begun to differ from values obtained at 1.06 [2] and 10.6 μm [1] and at dc. [9] The onset of this frequency dispersion in the avalanche breakdown process enables one to estimate the high-field electron-phonon collision frequency.

The laser system and techniques for avoiding and confirming the absence of self-focusing are described in detail in Refs. 2 and 10. Damage from inclusions was distinguished from intrinsic damage by examining both the morphology of the damage sites [1] and the temporal shape of light pulses transmitted through the sample. The latter technique employed the fact that a damaging light pulse is attenuated in a manner which is characteristic of the cause of damage. Only data obtained from intrinsic damage events were considered in the present work.

Figure 1 and Table I summarize the results of this study, the 1.06μm data from Ref. 2, and accepted dc results. [9] In figure 1 the damage fields of the various alkali halides are normalized to that of NaCl at the appropriate frequency. This makes the variation in trends between the 1.06 μm and the 0.69 μm measurements more easily seen. The relative breakdown strengths at 1.06 μm are virtually identical to those measured at dc. Although the corresponding data at 10.6 μm [1] are not displayed, they too follow the same trend. This is not the case at 0.69 μm, however, even when the 10 to 15 percent measurement errors are considered.

Table I lists the rms damage fields of NaCl for these experiments. The agreement found in the four experiments is heartening because there are particular difficulties in determining the absolute damage fields in dc measurements. [3] Root-mean-square values of the electric field strength are given because for laser pulses of this intensity, the buildup time of an electron avalanche to damaging levels is on the order of 10^3 to 10^4 cycles of the optical field. Heating of the electron population is thus effectively averaged over many cycles.

A complete theoretical description of avalanche breakdown has not been developed. The onset of frequency dispersion can be qualitatively understood, however, by using the results of models based on relaxation time approximations. [3, 11] These models predict that the damage field will scale with frequency as

$$E_{rms}(\omega) = (1 + \omega^2\tau^2)^{1/2} E_{dc} \quad (1)$$

where $\omega = 2\pi\nu$ is the laser radian frequency and τ is an effective, high-field, electron-phonon collision time. Since each material has a different value of τ, the relative breakdown fields for the alkali halides should begin to change at high frequencies. Data such as shown in Figure 1 can be used to infer approximate relative values of τ for a variety of theoretical models.

Perturbation calculations of τ have been performed which are applicable for electron energies greater than the longitudinal optical photon energies. [3] If $\omega\tau \approx 0.5$ for NaCl, then the results of these calculations can explain qualitatively the change in relative breakdown strengths observed at ruby frequencies for most of the alkali halides. The collision time for NaCl under this assumption is about 2×10^{-16} sec. For NaF these calculations predict that the relative breakdown field (E_{NaF}/E_{NaCl} in figure 1) will decrease at ruby frequencies, contrary to the change which is experimentally observed. This discrepancy may be the result of the inadequacy of the modeling of the electron-phonon interaction or it may indicate that the frequency dependence of the electron avalanche is not determined by the electron-phonon collision frequency alone.

Seitz [12] has suggested that the presence of deep-lying exciton bands may influence the dielectric strength of alkali halide crystals. If this is the case, then as the field frequency ω is increased, direct excitation out of these bands becomes possible and the damage field will decrease. NaF, which has the deepest lying bands (1.5-2.0 eV) of the materials studied, [13] will experience this effect at a higher frequency than the other alkali halides. Such considerations of the relative importance of the exciton bands may explain the observed large increase in the relative NaF damage field at 0.69 μm.

Multiphoton absorption directly across the bandgap cannot explain the changes in relative breakdown strength which have been observed. In addition, theoretical calculations of the fields at 0.69 μm necessary to induce damage from multiphoton ionization or from its low-frequency limit, tunnel ionization, give damage fields which are about an order of magnitude larger than those measured.

In this part of our work, we have measured the intrinsic optical breakdown fields of nine alkali halides using a ruby laser. Although the absolute and relative damage fields are comparable to the damage fields observed at 1.06 μm and at dc, differences are observed which suggest that at $\nu = 4.3 \times 10^{14}$ sec^{-1}, avalanche breakdown is no longer identical to dc avalanche breakdown. Current theories of avalanche breakdown do not appear to explain the details of this observed difference.

3. The Pulse-Duration Dependence of Optical Avalanche Breakdown

Measurements are reported here of optical damage induced by subnanosecond laser pulses. [4] These measurements were performed by focusing mode-locked YAG:Nd laser pulses having durations of 15 and 300 picoseconds inside single crystal NaCl. Because the experimental procedures used in the present work were identical to those used in the studies of Ref. 2 with a Q-switched YAG:Nd laser, the subnanosecond measurements can be directly compared to the results of those studies. It was found that the intrinsic breakdown field increased by almost an order of magnitude to over 10^7 volts/cm as the laser pulsewidth was decreased from 10 ns to 15 ps. The dependence of the damage field on laser pulse duration was used to calculate a field-dependent ionization rate. This rate was compared to that derived by Yablonovitch and Bloembergen from published values of the dc dielectric strength of NaCl [5] and good qualitative agreement found.

The laser used for the present work was a passively mode-locked Nd:YAG laser operating in a TEM_{00} mode at 1.06 μm. Without intercavity etalons, this oscillator produced bandwidth-limited light pulses of 15 picosecond duration. When the cavity output mirror was replaced with a sapphire etalon, the pulsewidth was lengthened to about 300 picoseconds. Two-photon-fluorescence measurements failed to detect substructure with pulses of either duration. A laser-triggered spark gap was used to select a single light pulse which, after attenuation, was focused through a 14 mm focal length lens about 2 mm into the sample. As in Ref. 2 care was taken to insure that spherical aberrations from both the lens and the plane entrance surface of the sample being tested were unimportant. An energy monitor recorded the energy in each laser pulse.

Although the intrinsic damage process in transparent materials is an inherently statistical process [6] (see Sect. 4) it is virtually threshold-like in NaCl. Consistent with Ref. 2, the damage field was defined as that value of root-mean-square electric field inside the sample which produced damage on a single shot with a probability of 0.5. Damage was identified by the occurrence of a faint spark and was accompanied by a small melted region ($\leq 2 \times 10^{-9}$ cm^3) inside the crystal. At least 20 data points were taken for each pulse duration at the 0.5 probability point.

Beam distortion from self-focusing was avoided by confining the laser input powers to well below the calculated critical powers for catastrophic self-focusing. (See Table II.) Tests discussed in Ref. 2 were used to confirm the absence of self-focusing. The absence of inclusion damage was confirmed in the manner discussed in Refs. 2 and 10.

Table II summarizes the results of the present measurements and relevant data from Ref. 2. An increase in breakdown strength was observed as the duration of the laser pulse was decreased. As the pulse duration was changed from 10.3 ns to 15 ps, there was a total change by a factor of 5.8 in damage field strengths or a factor of 33 in damage intensity. The experimental points are plotted in figure 2 along with the semi-empirical predicted curves from Ref. 5.

The existence of a pulsewidth dependence to intrinsic damage can be explained qualitatively by classical theories of electron avalanche ionization. [3, 11] These theories, which are summarized in Ref. 15, predict that the density of conduction-band electrons, N(t), increases with time as

$$N(t) = N_0 \exp \left[\int \alpha(e)\, dt \right] = M_c(t). \tag{2}$$

Eq. (2) is valid when, as is the case with laser breakdown, [5] electron diffusion and trapping can be ignored. N_o is the low density of conduction electrons before the application of the electric field and α is the ionization rate which increases monotonically with increasing electric field. Breakdown occurs when the density of electrons becomes high enough to cause a material irreversibility such as a phase change. As the time available for the avalanche to develop to damaging proportions decreases, the rate of ionization and hence the electric field must be increased in order to produce damage.

It is desirable to compare the laser data to dc results. Such a comparison cannot be made directly, because impulse dc measurements with subnanosecond impulse durations have not been made. As Yablonovitch and Bloembergen [5] have suggested, however, dc measurements on samples with varying thickness provide an indirect comparison because the maximum duration of the dc avalanche is limited to the electron drift time from cathode to anode. [3] By considering limits on the electron drift velocity, Yablonovitch and Bloembergen have calculated α (E) for dc fields using eq. (2) and previously published measurements of dc damage fields in thin samples of NaCl. [16] Breakdown was assumed to occur when M_c in eq. (2) reached a value of 10^8. The ionization rates for the laser data can be found by replacing the integral in eq. (2) by $\alpha(E_{rms})t_p$ where E_{rms} is the root-mean-square field on axis at the peak of the laser pulse and t_p is the laser pulsewidth. Then $\alpha(E_{rms})$ is given by

$$\alpha(E_{rms}) = \frac{1}{t_p} \ell n\, M_c \approx 18/t_p \tag{3}$$

This relation has been used to convert the quantity α (E) used along the vertical axis in the figure of Ref. 5 to our figure which used t_p^{-1}. We have shifted the curves along the horizontal axis to obtain agreement with the experimental values for the breakdown field E_{rms} for the long pulses.

In figure 2 the four laser measurements are plotted with the computed curve from Ref. 5. The two branches to the computed curve correspond to two limits on the high-field electron drift velocity. Within experimental error, the laser data overlap the upper curve of Ref. 5 which was derived on the assumption that the mobility in the hot electron gas is independent of E_{rms}. Quantitative agreement should not be emphasized, however, because the present analysis is based on at least two important assumptions which may not be valid over the range of damage fields considered.[15] The first assumption is that factors in the dc experiments such as space charges and electrode effects do not change as the sample thickness is reduced to approximately a micron. And the second assumption is that the same intrinsic mechanism dominates over the range of laser pulse widths in Table II. Another intrinsic mechanism -- multiphonon ionization [18] -- may cause damage at lower fields than required for avalanche breakdown when the laser pulsewidth is extremely short. Estimates for 1.06 μm radiation in NaCl indicate that when the laser pulsewidth is less than about a picosecond, multiphoton ionization is responsible for intrinsic damage.[5] Since the shortest pulsewidth considered in the present work is 15 ps, the neglect of multiphoton ionization appears to be justified. If the estimates of Yablonovitch and Bloembergen are inaccurate, however, and damage from multiphoton ionization is occurring, the ionization rate determined from the 15 ps pulse is an upper bound for the actual value of α at E_{rms} = 12.4 MV/cm.

Intrinsic laser-induced damage has been shown to be a pulse duration dependent process. As the laser pulse duration was decreased to 15 ps, the damage field in NaCl increased to over 10^7 v/cm. From the pulse duration dependence of the optical damage field, a field-dependent ionization rate was determined and found to agree at least qualitatively with experiments using dc fields. The agreement underscores the basic similarity between intrinsic laser-induced damage at 1.06 μm and dc electron avalanche breakdown.

4. Effects of Disorder on the Intrinsic Damage Field

Measurements are reported here of optical bulk damage in three disordered systems -- polycrystal KCl, a single-crystal KBr-KCl alloy, and fused quartz. In each case the damage field for the disordered system is compared to the optical strength of the corresponding crystal. These measurements were made in order to determine if the optical breakdown field increases with severe lattice disorder as had been observed in dc breakdown experiments [1, 19] and as predicted by simple theories of avalanche breakdown. [3]

The laser system and the experimental techniques used here were identical to those of Ref. 2 except that manner of attenuation of the transmitted laser light, rather than inspection of the damage was used to distinguish inclusion damage.[10]

It was found that the damage field of the large-grain (20 μm) polycrystal was the same as that measured in the single crystal and that the damage fields for the alloys were intermediate between the damage fields of the constituents. In quartz, on the other hand, the disordered (amorphous) form was noticeably stronger than the crystal, the ratio of damage intensities being 5 ± 1. This ratio is identical to the corresponding ratio of surface damage fields measured by Bass and Barrett.[6]

It is to be expected that the large-grain polycrystal should have the same damage field as the single crystal. The average grain diameter (20 μm) and the laser focal diameter are comparable so that in the high intensity region near the beam axis where breakdown is observed to initiate, the sample looks like a single crystal.

By a simplified argument we can predict the approximate crystallite size necessary to affect the breakdown strength. Classical theories of avalanche breakdown (see Ref. 15) predict that the dynamics of electrons with energies greater than the longitudinal optical (LO) energy determine the characteristics of the avalanche. The LO energy in the alkali halides corresponds to electron momenta of about 0.1 times the reciprocal lattice vector, G. Thus the important electrons have $k \geq 0.1\,G$. Phonons with values of $q \geq 0.1G$ will interact most strongly with these electrons. Because such lattice vibrations have wavelengths equal to 10 lattice constants or less, we expect that unless crystal disorder appears on the scale of about 10 lattice constants (~50A) or less, the damage field should be unaffected by disorder.

Amorphous systems may be disordered on such a scale. Our observation that fused quartz is more resistant to damage than crystalline quartz is, therefore, consistent with the argument just presented. This result can be explained in somewhat more quantitative terms. Eq. (1) shows that the rate of energy input into the electron population decreases with decreasing electron mobility, μ. In low electric fields the electron mobility in disordered media is less than in crystalline forms.[20] If the mobility in disordered media is also lower in the enormous fields characteristic of avalanche breakdown, then it should be more difficult to heat the electron distribution in fused quartz and, as observed it should be more damage resistant than the crystalline form.

The breakdown fields for various single crystal alloys of KBr and KCl are summarized in figure 3. It is seen that the damage strengths of the alloys are intermediate between those of the constituents. There is no evidence that alloying caused disorder sufficient to increase the breakdown strength of these materials. The variation of damage strength with composition can be qualitatively understood by noting that many material parameters such as bandgap, lattice constant, dielectric constant, and phonon frequencies have values intermediate between those of the constituents. According to simple models of avalanche breakdown, [3, 11] the breakdown field depends on these various material parameters so that it is reasonable to expect the breakdown strengths of the alloys to also be intermediate between those of the constituents.

It thus appears that only extreme lattice disorder such as present in highly disordered amorphous systems has a measurable effect on intrinsic damage fields. Further work is needed, however, to ascertain any general correlations between lattice disorder and breakdown fields. Such work is important not only to an understanding of avalanche breakdown but it is also important from a practical viewpoint to the design of more damage-resistant optics.

5. Laser Damage Statistics

In a recent study by Bass and Barrett [6] it was found that the resistance of surfaces to optical damage has a statistical character. This observation, which could apparently not be explained either by laser fluctuations or by fixed material inhomogeneities, was interpreted in terms of an electron avalanche model. Previously a number of investigators had measured a statistical character to dc breakdown in both gases [21] and solids. [22,23] Statistics in dc experiments were assumed to arise either from the details of an electron avalanche or from the dynamics of space charge formation.

The laser damage techniques of the present work can be applied to the study of breakdown statistics. There are, in fact, several advantages to using these techniques. First, the intrinsic damage process has been identified, at least in the alkali halides. Secondly, because damage from absorbing inclusions can be distinguished and space charges will presumably not develop in optical fields, major experimental uncertainties in the surface studies and dc measurements are avoided. And finally, the bulk of crystals is far better characterized than surfaces in terms of structure and composition.

We present new experimental evidence to support the conclusion that there exists a statistical nature to laser-induced damage both in the bulk and on the surfaces of transparent materials. [10] It will be shown that the new experimental data is compatible with the results of Ref. 6.

It was observed in Ref. 6 that a precisely defined threshold for laser induced surface damage does not exist in the ten different solids which were studied. Instead, there is a range of power levels within which damage can develop on each shot with some finite probability, p_1, such that $0 < p_1 \leq 1$. The damage probability, p_1, at some power level was defined as the ratio of the total number of damage sites to the total number of laser shots. It was found that p_1 as a function of the electric field, E, appears to satisfy the relationship

$$p_1 \propto \exp(-K/E). \tag{3}$$

Additional evidence for the probabilistic nature of laser damage was obtained in experiments in which an image converter streak camera was used to measure the distribution of breakdown starting times for surface damage to two different materials. [24]

During the Q-switched damage experiments on the alkali halides, evidence was recorded which supports the statistical viewpoint. By monitoring the transmitted laser light with a fast photodiode-oscilloscope combination (risetime ≅ 0.5 ns), we observed that the laser light is attenuated when damage develops. The first instant of measurable attenuation can occur before, at, or after the peak of the laser pulse, so that no well-defined relationship exists between the laser intensity and the first instant of attenuation. Figure 4 shows examples of such observations made with a ruby laser beam focused to produce damage inside NaCl. The laser pulses are fully time-resolved as verified by Fabry-Perot studies. In figure 5 another ruby laser pulse was focused into NaCl but did not cause damage whereas a second pulse, apparently identical to the first, did induce damage when focused into the same volume of the crystal. Nothing was moved between the two laser shots, and the laser was firing automatically at a repetition rate of about 1 pulse/5 sec. Similar observations were recorded at 1.06 μm where the automatic firing rate was just over 1 pps.

The relationship between laser light attenuation and the size of the electron avalanche is difficult to establish. A reasonable estimate indicates that the transmitted light is unaffected by the avalanche until the density of conduction-band electrons reaches a level of about 10^{18} cm^{-3}. It is not strictly correct, therefore, to associate the first instant of attenuation with the breakdown or avalanche starting time. We will assume, however, as a simplification that the two instants of time are identical within experimental error. It should be noted, incidentally, that the time interval over which the transmission drops from effectively unity to nearly zero may have no simple relationship to the ionization rate, $\alpha(E)$, defined in Sect. C. Because the density of electrons is quite high by the time measurable attenuation occurs, electron-electron collisions may alter the avalanche development and reflection from the plasma may become important, particularly at longer laser wavelengths.

Encouraged by our experimental observations, we decided to conduct careful measurement of the statistical time lag in a number of materials. Figures 6 and 7 summarize measurements made in fused quartz at 1.06 μm, in NaF at 0.69 μm. Similar results were obtained for sapphire at 1.06 μm. As usual, self-focusing was found to be absent under the conditions of our measurements. Techniques discussed in Refs. [2] and [10] were used to verify that the statistics were not occurring because of the presence of point-to-point material inhomogeneities such as inclusions.

The distributions of breakdown starting times for surface and bulk damage to fused quartz are shown in Fig. 6 to be virtually identical for the same value of p_1. These distributions have the same characteristics as those reported by Bass and Barrett for entrance surface damage. [24] The breakdown can begin at any time over a relatively long interval which includes times after the peak intensity in contrast to the very sharply defined breakdown starting time expected for a threshold-like process. The most probable time for damage is, in all cases, before or at the instant of maximum intensity or optical field, and as the applied field is reduced (p_1 is lowered), the time of maximum damage probability shifts to the peak of the pulse. (Compare Figures 6c and d.) These qualitative properties were shown in Refs. 10 and 24 to be explained by the probabilistic point of view.

In Ref. 10 a detailed comparison is made between the results of Ref. 6 and the present data. Figure 8 summarizes the important results of that comparison. A computed distribution of bulk breakdown starting times in fused quartz, obtained from Eq. (3) and appropriate measured parameters is compared to the observed distribution for bulk damage in fused quartz. [10] Good qualitative agreement is obtained. For times after the peak of the laser pulse, there is some quantitative discrepancy between the computed curve and the experimental results. As discussed in Ref. 10, this discrepancy appears to be the result of the finite formation time for the avalanche.

The present work confirms the existence of a statistical character to the laser induced intrinsic damage process and supports the notion that the intrinsic damage mechanism both on the surface and in the bulk is an electron avalanche with statistical starting times.

6. Optical Surface Damage

In the experiments of Bass and Barrett, [6] the laser beams were focused to about 25 μm in diameter ($1/e^2$ in intensity) in order to induce damage on the surface. For such focal diameters the laser beam was effectively collimated over a distance of about 50 μm near the focus, and yet damage was consistently observed to develop within the first 0.25 μm of material. Because surface contaminants were apparently not responsible for damage in this study, this surprising result indicated that the surfaces of conventionally polished solids have a lower intrinsic threshold than the bulk. The same conclusion follows from a later work by Giuliano [25] in which it was observed that the surface damage field for sapphire was increased when the surface was ion-beam polished, a procedure that removed most but not all of the polishing scratches on his samples.

A quantitative relationship between surface damage fields and those of the bulk was not established experimentally in these or in any earlier study, because until the work of Yablonovitch, [1] no accurate measurements of intrinsic optical bulk damage had ever been made. Using the bulk damage techniques of Ref. 2 and the surface focusing scheme of Ref. 6, we have completed a study of surface damage at 1.06 μm in which surface damage fields were measured for a variety of surface preparations and compared to bulk damage fields. Because this work has already appeared in the literature, [26] , only a brief summary of the results will be given.

The surface breakdown fields, E_S, were determined for various materials whose surfaces had been prepared by three techniques: conventional abrasive polishing, a bowl-feed technique developed by Itek Corp., and Ar-ion beam polishing. For each sample, a measurement of the bulk intrinsic damage field, E_B, provided a direct comparison and reference.

Table III summarizes the measured ratios E_B/E_S and Table IV lists the measured values of E_B in the three materials studied. These data show that the clean but conventionally polished surface of a transparent medium is generally more easily damaged than the bulk. On the other hand, when care is taken to achieve imperfection-free surface finishes by the use of bowl-feed or ion polishing, the bulk and damage fields are equal.

Bloembergen has shown that the discontinuities in the dielectric constant occurring at structural defects can enhance the electric field over its average value. [7] The following expressions for the ratios E_B/E_S when small surface defects are present were given in Ref. 7:

spherical void	$3\epsilon/(2\epsilon + 1)$
cylindrical groove	$2\epsilon/\epsilon + 1)$
"Vee" groove	ϵ

where ϵ is the optical dielectric constant. These were used to obtain the predicted ratios in Table III.

The mechanical defects that result from standard abrasive polishing [27] are responsible for the agreement between the predicted and experimentally measured ratios for conventionally finished surfaces. The bowl-feed and ion-beam polished samples appear to have only gentle variations in surface topography [8] which should lead to little if any measurable field enhancement. In agreement with this idea we found no difference between surface and bulk breakdown fields for these samples.

It should be noted, incidently, that good quality conventional polishes appear spatially uniform to a 20 μm-diameter laser beam and so the distribution of defects does not cause damage statistics. Intrinsic damage on such a surface will produce damage statistics of the form of Figs. 6 and 7 in Sect. 5 .

This work has shown that the incident laser fields for intrinsic entrance surface and bulk damage to transparent solids are the same as for imperfection-free surfaces. When structural imperfections are present, they enhance the optical field locally so that the surface damages more easily than the bulk. The present results and those of Sect. 5 demonstrate that the intrinsic mechanisms for optical damage to surfaces and to the bulk of transparent solids are the same, namely electron avalanche breakdown.

7. Implications of Experimental Results to Theory

A complete theory of avalanche breakdown has not been developed. The basis of the difficulty in modeling the avalanche is the fact that the relevant interactions cannot be treated separately. It is not strictly correct, for example, to consider electron excitations as distinct from those of the lattice. The electron-phonon interactions in the alkali halides are so strong that effective collision times appear to be about 2×10^{-16} sec. Such short times imply an electron energy uncertainty of the order of the ionization energy. In addition, lattice distortions from electric fields having rms values of 10^6 V/cm or more cannot properly be treated as a perturbation on the energy spectrum of the electron-lattice spectrum.

Our present experimental results provide valuable data for understanding aspects of the avalanche process. In addition to measuring accurate breakdown fields, we were able to infer an approximate ultra-high-field electron-phonon collision time in NaCl, a parameter which cannot be obtained by other experiments. We have, in addition, confirmed that the avalanche breakdown mechanism is pulse duration dependent. This latter conclusion refutes a number of avalanche models which view breakdown as the result of a discontinuous jump in electron density as the external field is raised. [3]

The data on the pulse-duration dependence of breakdown fields has been used to infer an ionization rate α (E) which in turn can be compared to the predictions of simple models of breakdown. One such model is that of Shockley [26] who found that the ionization rate α (E) should have the functional

form A exp (-B/E) where A and B are appropriate constants. A comparison to Shockley's model is made in Fig. 9 where it is seen that Shockley's result is not an unreasonable approximation to the experimental data.

The high value of effective electron-phonon collision frequency $1/\tau$ can be shown [15] to imply that the electron-phonon interaction appropriate to a description of breakdown in insulators is not described by the Frohlich Hamiltonian as widely assumed. [3,11] A deformation or non-polar interaction discussed many years ago by Seitz [12] may be more useful in describing the effective electron-lattice interaction in avalanche breakdown than is the Frohlich interaction.

Our measured values of breakdown fields can be shown to imply that the average electron energy is much less than the ionization energy when the avalanche occurs. [15] This observation lends support to the conclusions of Baraff [29] and Holway [11] who have found, using simplified classical models, that the diffusion of electrons towards the high energy tail of the energy distribution appears to be more important than the average electron energy in determining the avalanche characteristics.

Finally, we have seen that severe crystal disorder can increase the damage fields for solids and that the avalanche process has a statistical character. Both observations can be shown to support the identification of the damage process as being an electron avalanche [6,15]. The measured characteristics of the damage statistics may, in addition, provide valuable insight into the physical development of the avalanche.

In conclusion, although a complete theory of avalanche breakdown may be many years distant, we can understand many features of our experimental results in terms of relatively simple models. Experiment must lead theory in this area, and data gathered in work such as we have reported here may provide both a basic foundation for and a test of future theories of avalanche breakdown.

8. Summary

The intrinsic optical damage process in transparent solids has been shown to be electron avalanche breakdown. This process has been a frequency and pulse-duration dependence. Severe crystal disorder can increase the breakdown strength of materials, and the damage process itself has been shown to have a measurable statistical aspect.

Such results are important for the theoretical understanding of avalanche breakdown. They also provide measured upper limits for the propagation of high intensity light in solids and indicate how these limits change with laser pulse and material characteristics. By using techniques appropriate to the study of intrinsic damage, we have been able to clarify the relationship between optical surface and bulk damage and to gather information which may be useful in the development of more highly damage resistant materials and coatings.

9. Acknowledgments

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10. References

[1] Yablonovitch, E., Appl. Phys. Lett. 19, 495 (1971).

[2] Fradin, D. W., Yablonovitch, E., and Bass, M., Appl. Optics 12, 700 (1973).

[3] O'Dwyer, J. J., The Theory of Dielectric Breakdown of Solids (Oxford University Press, London), (1964).

[4] Conwell, E. M., High Field Transport in Semiconductors, Solid State Physics Supplement 9 (Academic Press, New York and London), (1964).

[5] Yablonovitch, E. and Bloembergen, N., Phys. Rev. Lett. 29, 907 (1972).

[6] Bass, M., and Barrett, H. H., IEEE J. Quant. Elect. QE-8, 338 (1971).

[7] Bloembergen, N., Appl. Optics 12, 661 (1973).

[8] Fradin, D. W. and Bass, M., Appl. Phys. Lett. 22, 206 (1973).

[9] von Hippel, A., J. Appl. Phys. 8, 815 (1937).

[10] Bass, M. and Fradin, D. W., IEEE J. Quantum Elect., to be published (Sept. 1973).

[11] Holway, L. Jr., Phys. Rev. Lett. 28, 280 (1972).

[12] Seitz, F., Phys. Rev. 76, 1376 (1949).

[13] Teegarden, K., and Baldini, G., Phys. Rev. 155, 896 (1967).

[14] Fradin, D. W., Bloembergen, N., and Letellier, J. P., Appl. Phys. Lett., to be published (15 June 1973).

[15] Fradin, D. W., Final Report for Joint Services Electronics Program at Harvard University under Contract No. N00014-67-A-0298-0006 (May 1973).

[16] Watson, D. W., Heyes, W.,Kao, K.C., and Calderwood, J. H., IEEE Trans. Elec. Insul. EI-1, 30 (1965).

[17] Prof. Y. R. Shen has suggested (private communication, 1972) that local field effects may be important in an electron avalanche and may explain the difference in absolute field strengths between dc and optical frequencies. The damage fields reported in the literature, however, are not corrected for local fields because it is normally assumed that any local field effects are averaged out by the electrons' rapid movement across the unit cell. The validity of this latter assumption has not been established.

[18] Slusher, R.E., Giriat, W., and Brueck, S.R.J., Phys. Rev. 183, 758 (1969).

[19] von Hippel, A., and Mauer, R. J., Phys. Rev. 59, 820 (1941).

[20] Kittel, C., Introduction to Solid State Physics, Third Edition (John Wiley and Sons, New York), (1967).

[21] Wijsman, R. A., Phys. Rev. 75, 833 (1949).

[22] Kawamura, H., Ohkura, H., Kikuchi, T., J. Phys. Soc. Japan 19, 541 (1954).

[23] Watson, D.W., Heyes, W., Kao, K.C. and Calderwood, J. H., IEEE Trans. Elect. Insul. EI-5, 58 (1970).

[24] Bass, M., and Barrett, H.H., Applied Optics 12, 690 (1973).

[25] Giuliano, C.R., Appl. Phys. Lett. 21, 39 (1972).

[26] Fradin, D.W. and Bass, M., Appl. Phys. Lett. 22, 157 (1973).

[27] Khan, J. M., private communication (1972).

[28] Shockley, W., Czech. J. Phys. B11, 81 (1961) and Sol. St. Elect. 2, 35 (1961).

[29] Baraff, G.A., Phys. Rev. 128, 2507 (1962).

Table 1. Absolute Breakdown Strength of NaCl

E_{peak} (dc)*	1.50×10^6 V/cm
E_{rms} (10.6 μm)*	$(1.95 \pm 0.20) \times 10^6$ V/cm
E_{rms} (1.06 μm)*	$(2.3 \pm 0.46) \times 10^6$ V/cm
E_{rms} (0.69 μm)	$(2.2 \pm 0.44) \times 10^6$ V/cm)

*These values are taken from Refs. 9, 1, and 2 respectively.

Table 2. Experimental Breakdown Fields and Calculated Self-Focusing Parameters in NaCl

Pulsewidth (10^{-12} sec)	P_{input} (10^6 watts)	P_c (10^6 watts)* electrostriction	electronic	E_{rms} (10^6 volts/cm) relative	absolute
15	1.5	2.9×10^4	18	E(15ps)/E(300ps)	12.4 ± 3.7
300	0.22	82	18	$= 2.6 \pm 0.7$	4.7
4.7×10^3	0.030	1.8	18	E(4.7ns)/E(10.3ns)	2.3 ± 0.4
10.3×10^3	0.033	1.8	18	$= 1.1 \pm 0.05$	2.1

*P_c is the calculated critical power for catastrophic self-focusing

Table 3. Comparison of Bulk and Surface Damage Fields for Different Samples and Surface Finishes

Material	Finishing Procedure Finish Quality[a]	Predicted Ratio E_B/E_S Spherical Void	Cylindrical Groove	"Vee" Groove	Measured E_B/E_S	Surface Damage Morphology
Fused Quartz	Conventional #1 Standard 0-0 (see Fig. 1a)	1.21	1.35	2.1	1.3±0.1	b
	Conventional #2 Standard 0-0	"	"	"	1.5±0.1	b
	"Bowl" feed finish See Fig. 1b	"	"	"	1.0±0.1	c
	Ion beam polish Standard 0-0 with final 1.25 1.25 μm removed by Ar ion beam	"	"	"	1.0±0.1	c
Sapphire	Conventional Many scratches and digs with 50X mag and dark field	1.29	1.50	3.00	2.0±0.2	b
BSC-2 Glass	Conventional Standard 0-0	1.23	1.39	2.25	1.3±0.1	b
	"Bowl" feed finish (see Fig. 1c)	"	"	"	1.0±0.1	b, c

a. All surfaces were cleaned with collodion

b. Very faint pit ~ 20 μm in dia. and ~ 0.25 μm deep

c. Extensive cracking out to ~ 150 μm with central hole ~ 20 μm dia. and ~3-10 μm deep

Table 4. Measured Intrinsic Bulk Damage Fields at 1.06 μm

	Material	E_B(MV/cm)[a]
Fused Quartz	Conventional #1	5.2 ± 1.3
	Conventional #2	
	Bowl feed finish	
	Ion beam polishes	
Sapphire		6.3 ± 1.6
BSC-2 Glass		4.7 ± 1.2

(a) RMS field values are listed
Measurements were made relative to E_B in NaCl
Pulse duration ~ 10 nsec (FWHM)

Figure Captions

Fig. 1 RMS Electric Fields Necessary to Induce Damage in Nine Alkali Halides Normalized to the Damaging Field for NaCl. The dc data is taken from Ref. 10 and 1.06 μm is taken from Ref. 7.

Fig. 2 The Rate of Ionization in NaCl as a Function of Electric Field
The two branches of the solid curve correspond to two limits on the high-field electron drift velocity which were used in Ref. 5 to calculate the dc ionization rate. Corresponding laser data at 1.06 μm is plotted. It should be noted that the changes in the damage field as the pulse duration is lowered from 300 to 15 ps and from 10.3 to 4.7 ns are known to higher precision than the absolute field strengths. The error bars reflect the experimental uncertanties in the absolute field strengths.

Fig. 3 Measured Relative 1.06 μm Breakdown Fields for KCl-KBr alloys

Fig. 4 The Occurrence of Internal Damage in NaCl Due to Ruby Laser Irradiation. The laser intensity transmitted through the sample is shown in these photos.

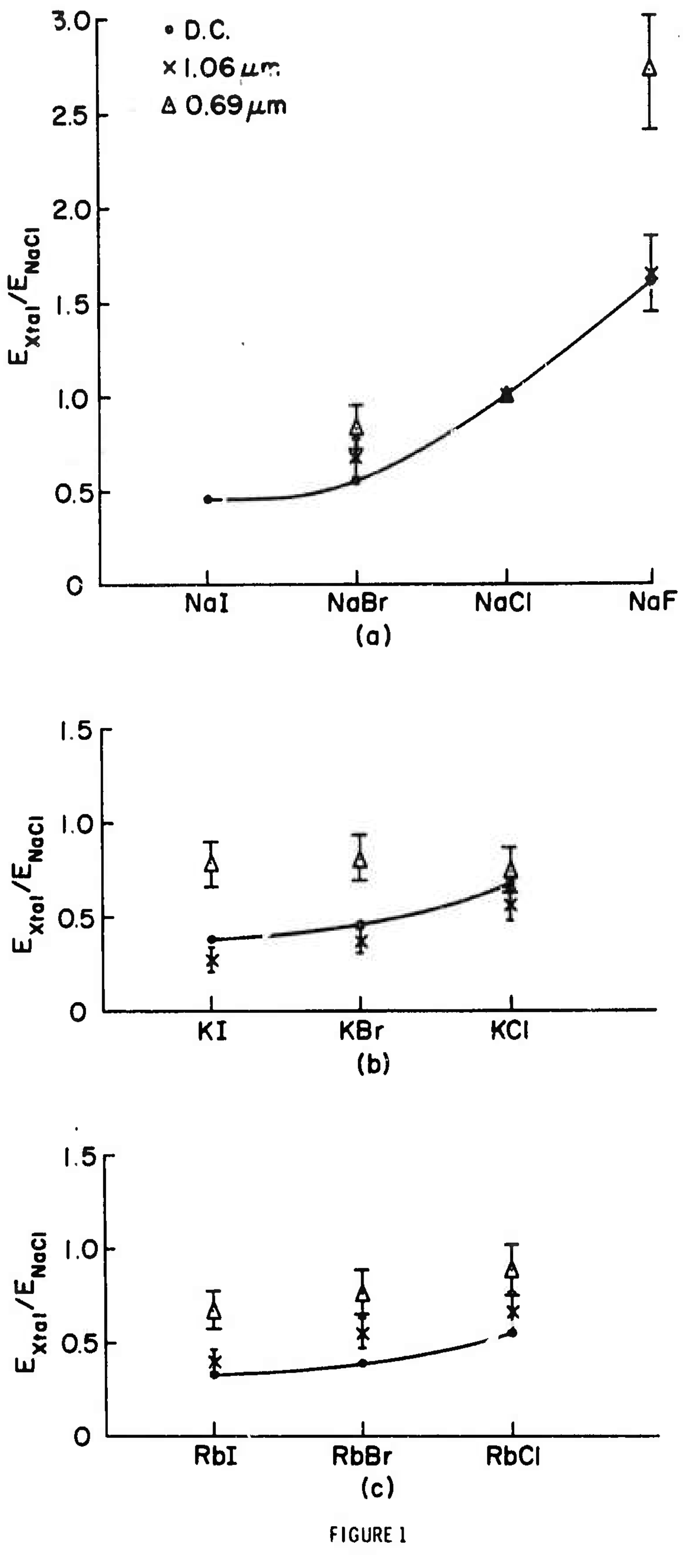
Fig. 5 Intrinsic Bulk Damage Occurring in NaCl on the Second of Two Ruby Pulses. Five seconds passed between pulses.

Fig. 6 Measured Distributions of Breakdown Starting Times for Fused Quartz Using Nd:YAG Laser Irradiation

Fig. 7 Measured Distribution of Breakdown Starting Times for NaF Using Ruby Laser Irradiation

Fig. 8 Measured and Calculated Distributions of Breakdown Starting Times for Bulk Damage in Fused Quartz using a Nd: YAG Laser.

Fig. 9 The linear plot is the function exp $(-K/E)$ where $K = 17$ MV/cm. It is seen that this function is a reasonable fit to the experimental data. The ionization rate α is related to the pulse width by Eq. 3.



• D.C.
× 1.06 μm
Δ 0.69 μm
E_{Xtal}/E_{NaCl}
3.0
2.5
2.0
1.5
1.0
0.5
0
NaI
NaBr
NaCl
NaF
(a)
1.5
1.0
0.5
0
KI
KBr
KCl
(b)
1.5
1.0
0.5
0
RbI
RbBr
RbCl
(c)

FIGURE 1

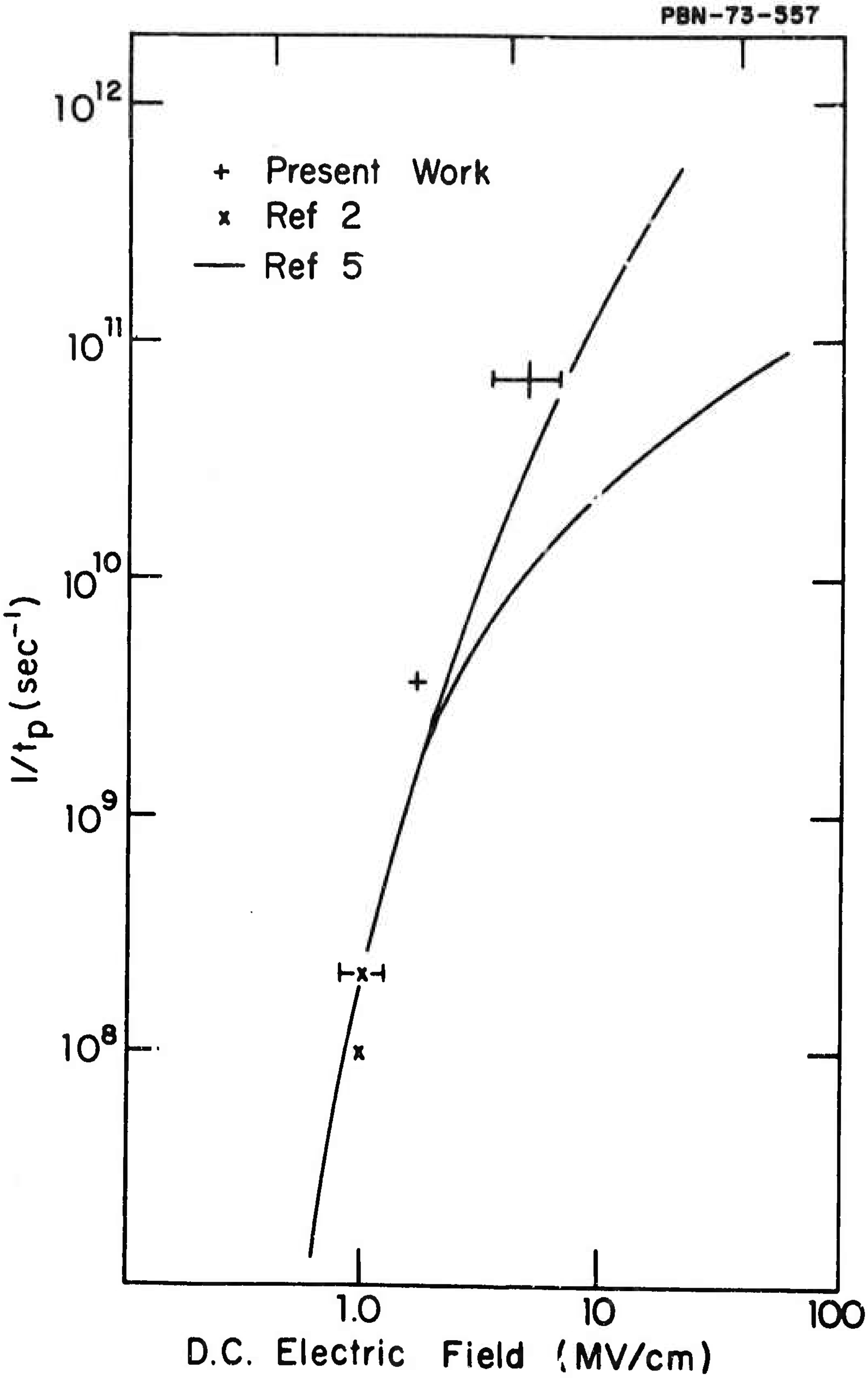
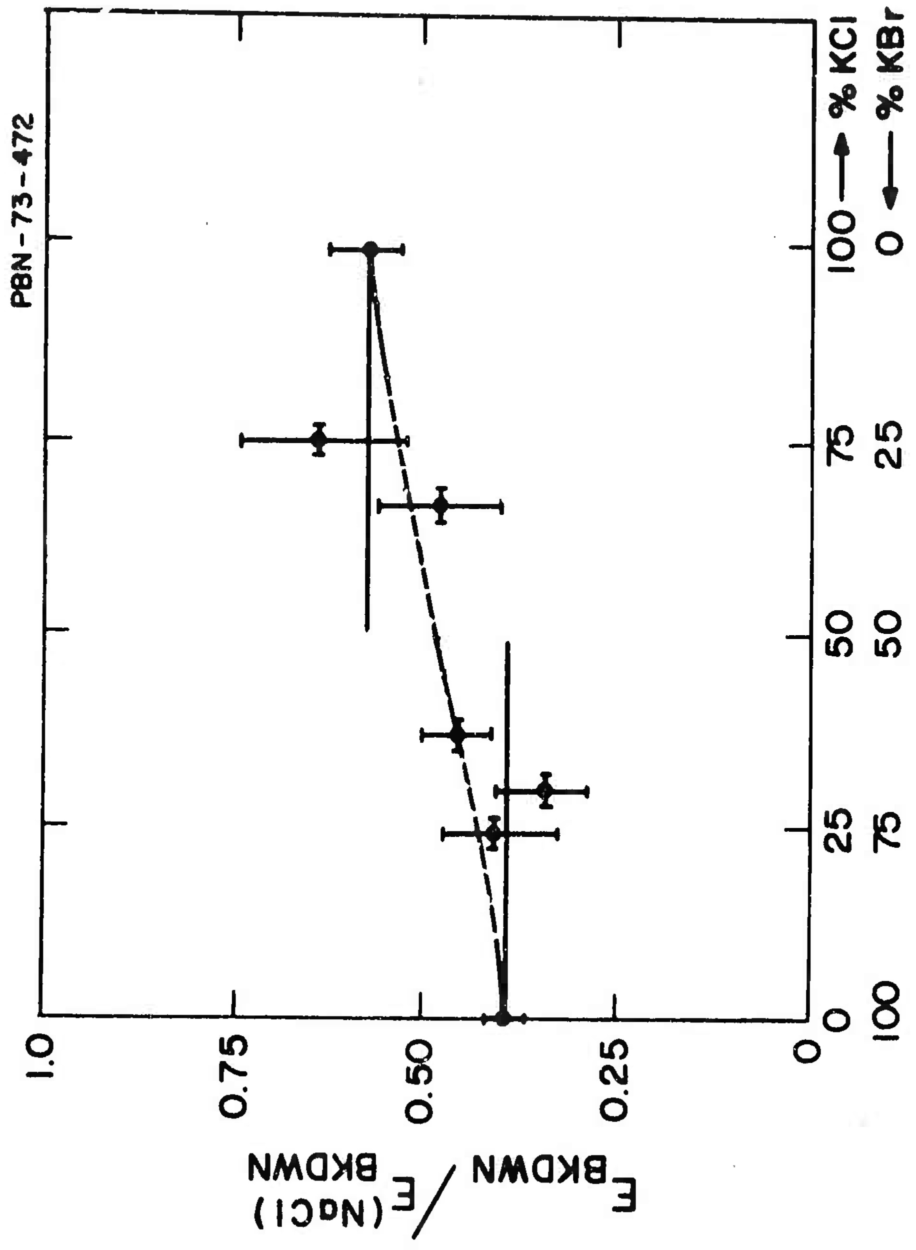
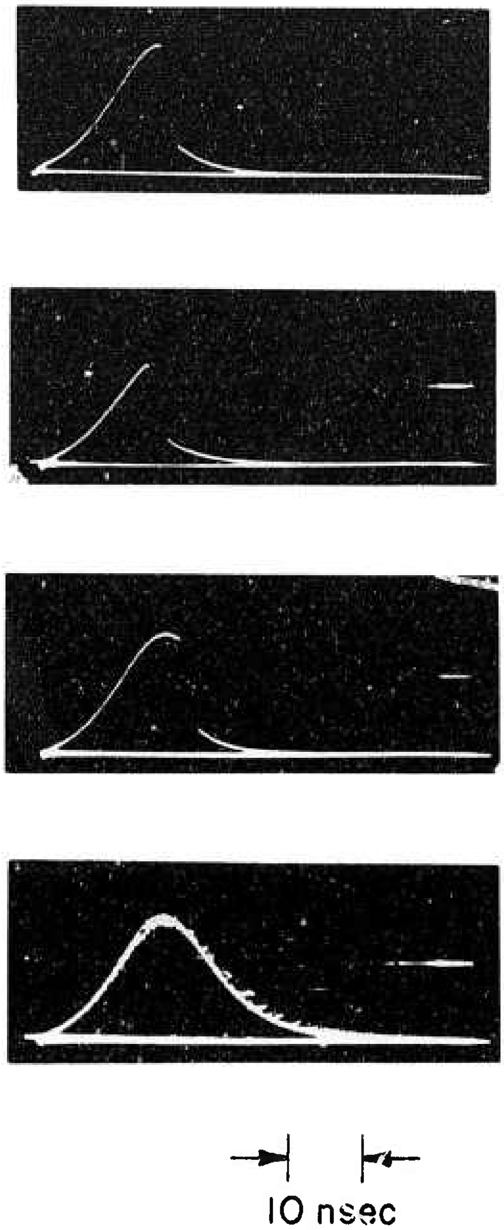

PBN-73-557
10^{12}
10^{11}
10^{10}
10^{9}
10^{8}
$1/t_p$ (sec^{-1})
+ Present Work
x Ref 2
— Ref 5
1.0
10
100
D.C. Electric Field (MV/cm)

FIGURE 2



PBN-73-472
1.0
0.75
0.50
0.25
0
$E_{BKDWN} / E_{(NaCl)\,BKDWN}$
0 25 50 75 100 → % KCl
100 75 50 25 0 ← % KBr

FIGURE 3



a) Damaged when the peak laser field was reached $E_{Damage}/E_{Peak} = 1$

b) Damaged before the peak laser field was reached $E_{Damage}/E_{Peak} = 0.896$

c) Damaged after the peak laser field was reached $E_{Damage}/E_{Peak} = 0.954$

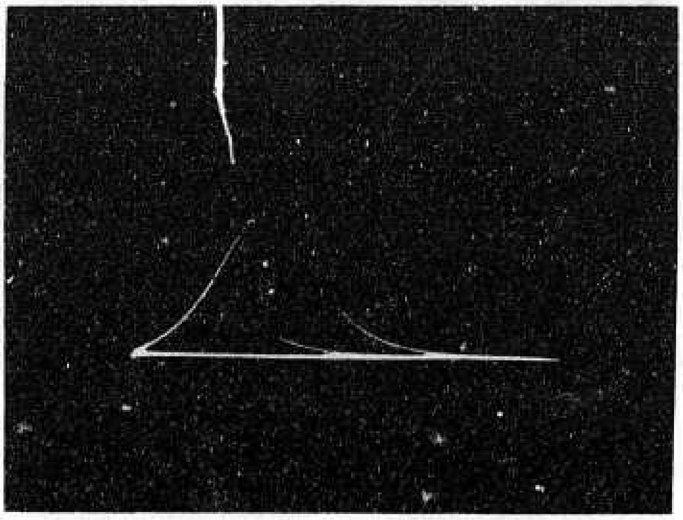
d) 3 successive pulses no damage $E_{Peak} \equiv 1$ (Arbitrary units)

<u>THE OCCURRENCE OF INTERNAL DAMAGE IN NaCl</u>

A TEM_{00} mode ruby laser with total pulse energy of 0.3 mJ was focused inside the inclusion free sample with a 14mm focal length lens

FIGURE 4

PBN-72-543



→| |← 10 nsec

Ruby laser pulses in NaCl with no self-focusing. The first pulse caused no damage. Five seconds later the second pulse caused damage before peak field was reached.

FIGURE 5

PBN-72-1172

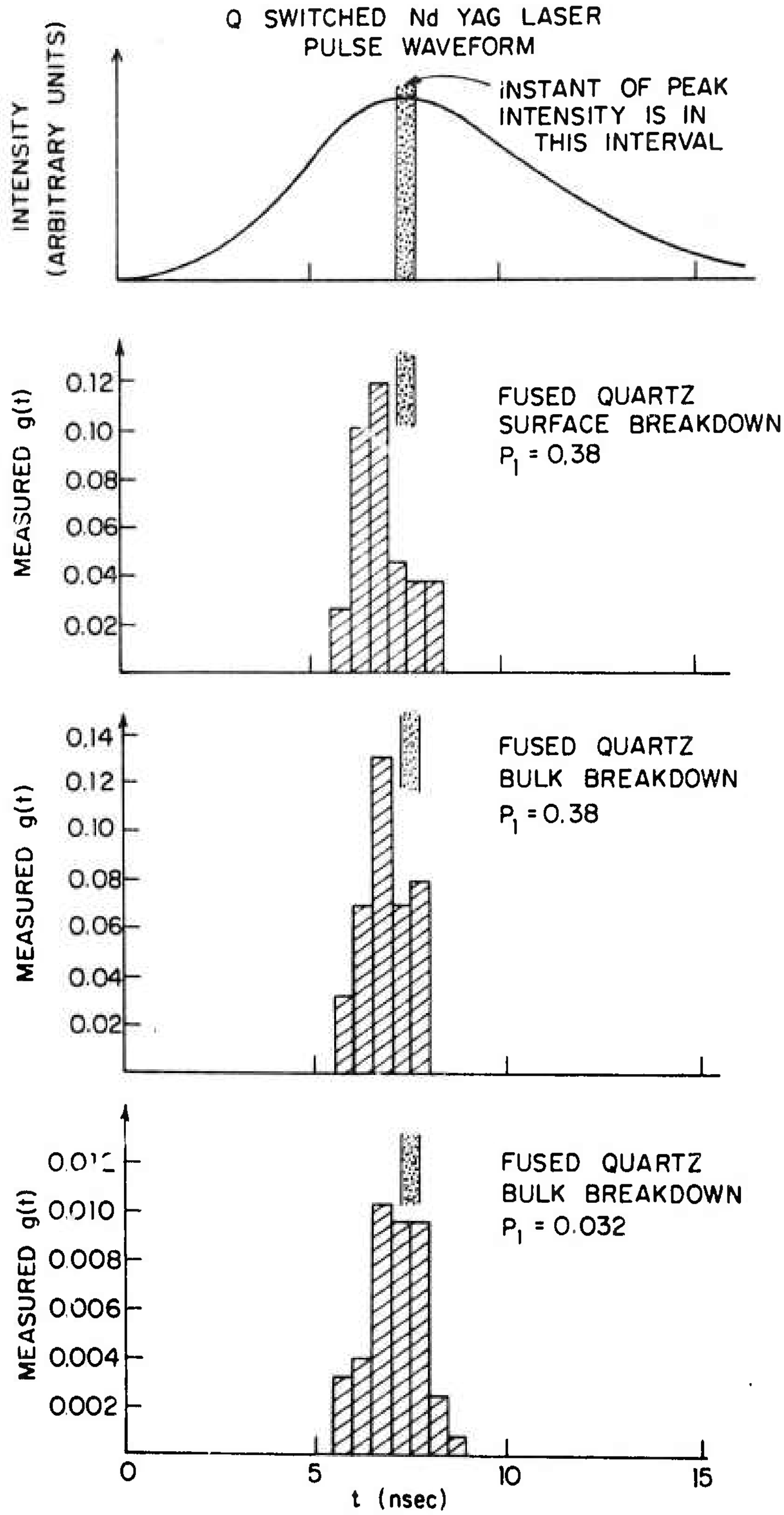


FIGURE 6

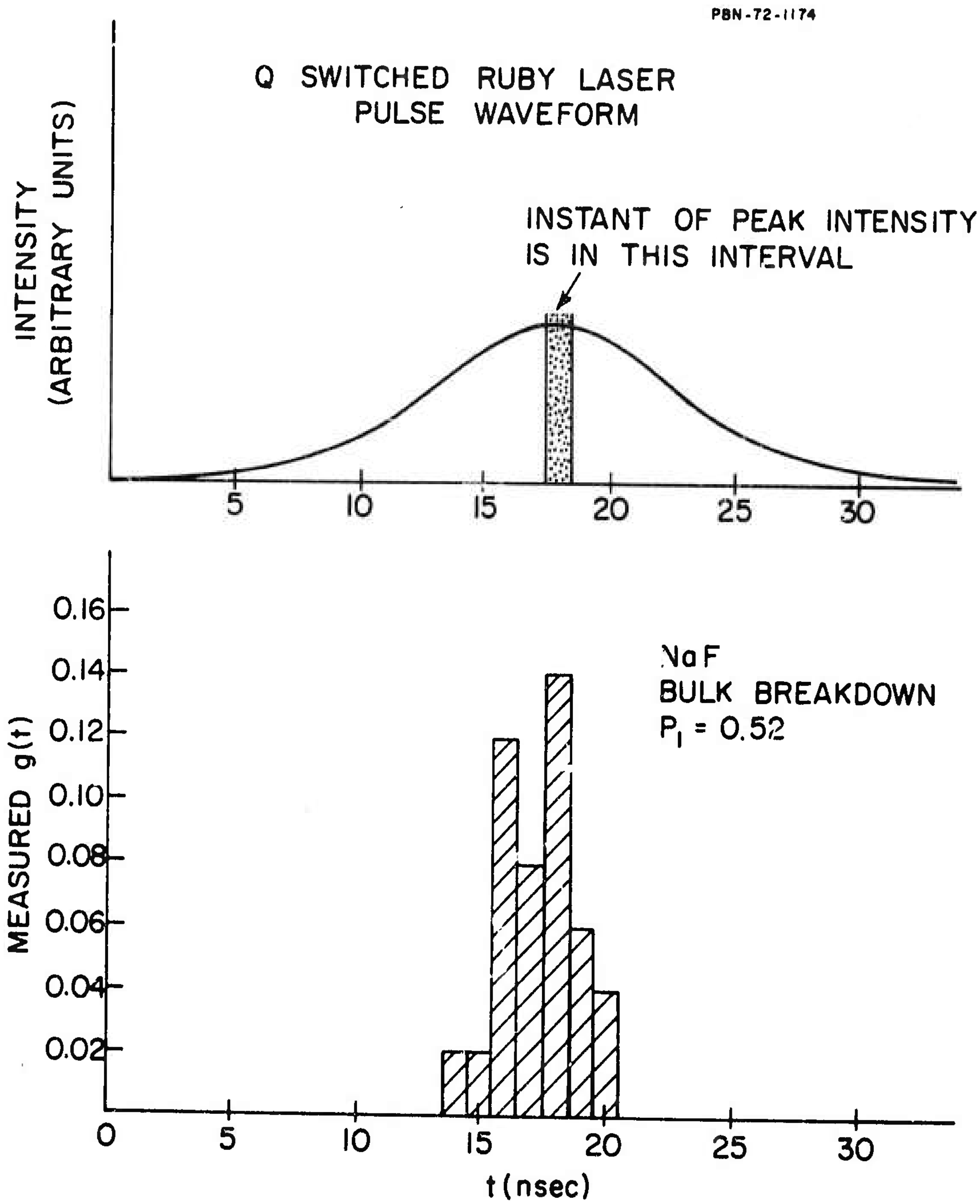

PBN-72-1174
Q SWITCHED RUBY LASER
PULSE WAVEFORM
INTENSITY
(ARBITRARY UNITS)
INSTANT OF PEAK INTENSITY
IS IN THIS INTERVAL
5
10
15
20
25
30
0.16
0.14
0.12
0.10
0.08
0.06
0.04
0.02
MEASURED g(t)
NaF
BULK BREAKDOWN
P1 = 0.52
0
5
10
15
20
25
30
t (nsec)

FIGURE 7

PBN-72-1175

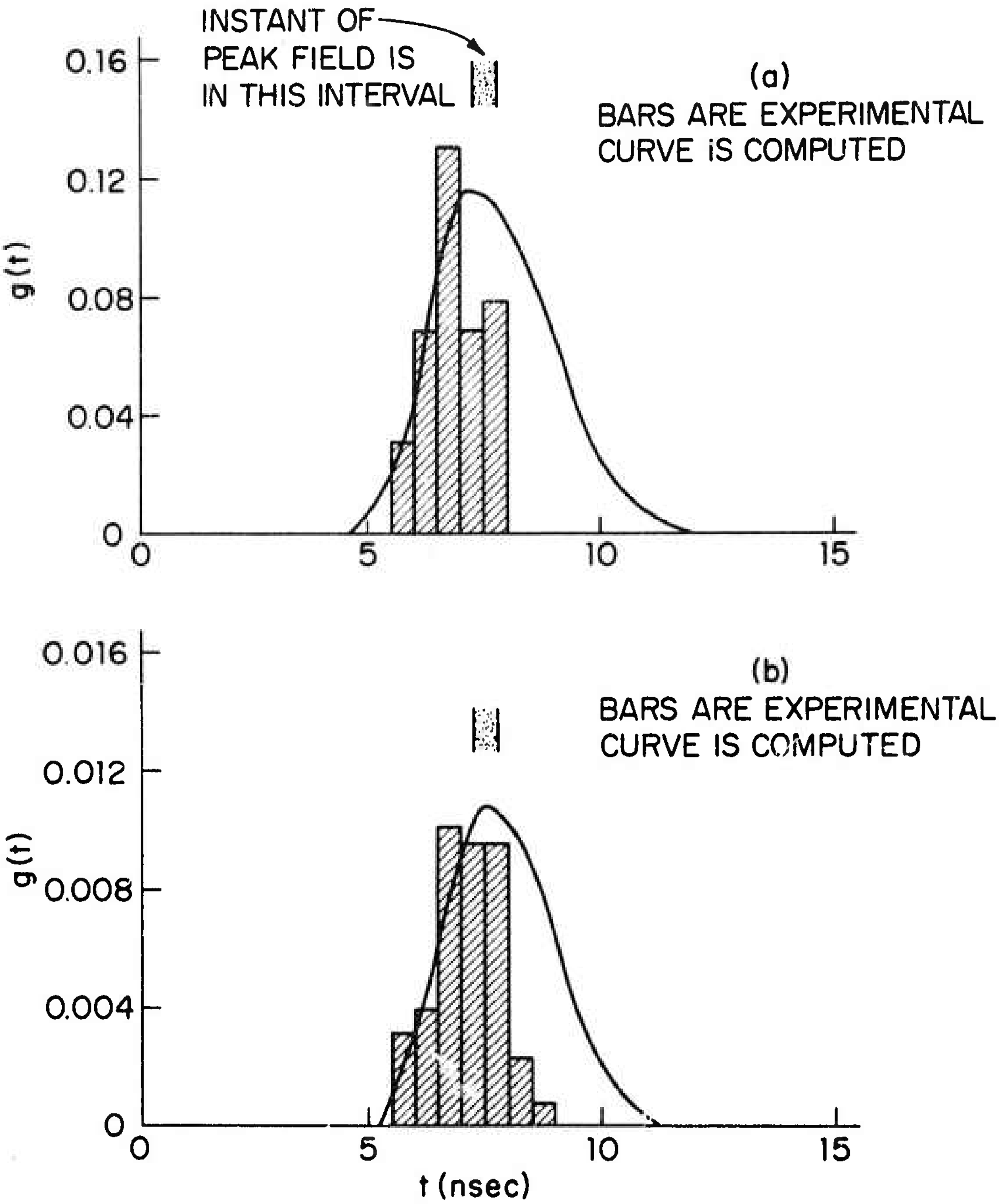


FIGURE 8

PBN-73-556

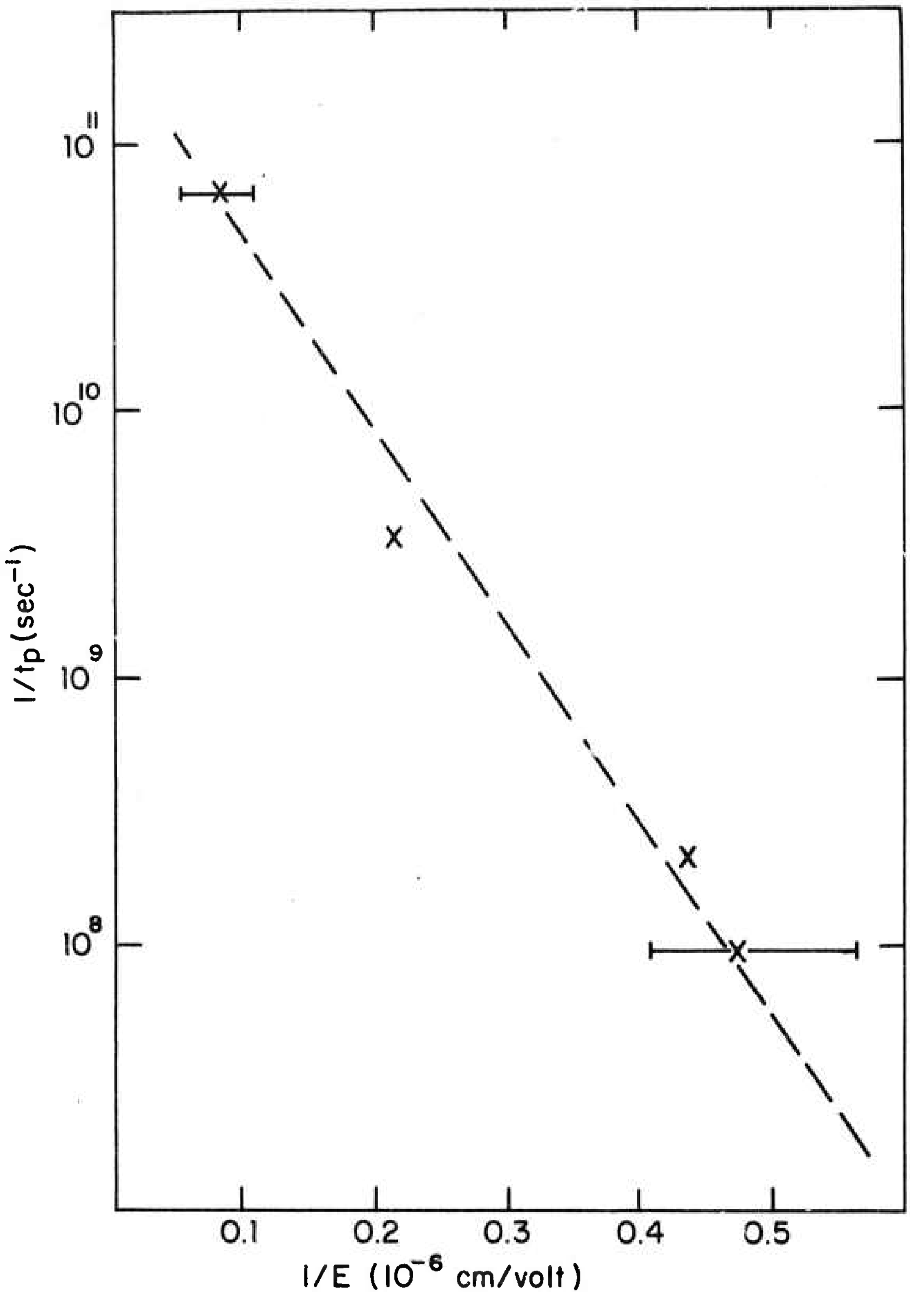


FIGURE 9